

# Mode Coupling Approach for spin-facilitated kinetic Ising models

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The  $d$ -dimensional  $n$ -spin facilitated kinetic Ising model is studied analytically starting from usual master equations and their transformation into a Fock-space representation. The evolution of relevant operators is rewritten in terms of a projection formalism. The obtained frequency matrices and memory terms are analyzed. Especially, the influences of the memory terms is approached by using standard techniques of the usual mode coupling approach. The temperature dependence of the relaxation times related to the  $n$ -spin facilitated kinetic Ising model shows a weak non-Arrhenius behavior. Furthermore, a characteristic stretched decay of the correlation function is obtained.

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## I. INTRODUCTION

In spite of advances in the description of liquids near the glass transition using different approaches [1–4] the phenomenon is generally not completely understood. Supercooled fluids reveal often a non-Arrhenius behavior of relaxation times and a characteristic stretched exponential decay of correlation functions. But a long range order is not developed in contrast to conventional phase transitions. The dynamics of the glass transition can be described by an increasing cooperativity of local processes with decreasing temperature [5]. The cooperativity leads to the well known slowing down of the relaxation times which can be illustrated by a strongly curved trajectory in the Arrhenius plot (logarithm of the relaxation time  $\tau$  versus the inverse temperature  $T^{-1}$ ). One possible fit of this curve is given by the Williams-Landel-Ferry (WLF) relation [6], i.e.  $\ln \tau \propto (T - T_0)^{-1}$  with a finite Vogel temperature  $T_0$ .

Mode coupling theories [1,7,8] (MCT) predict a completely ergodic decay of correlation functions above a critical temperature  $T_c$  and an incomplete decay below  $T_c$ . The incomplete decay is usually identified as the relatively fast  $\beta$ -process caused by processes related to the breaking up of the local cages. The long time regime of the ergodic decay above  $T_c$  is denoted as  $\alpha$ -process. The relaxation time of the  $\alpha$ -process increases rapidly with decreasing temperature and below  $T_c$  only the  $\beta$ -process remains effective, i.e. at  $T_c$  the system undergoes a sharp phase transition to a state with partially frozen (density) fluctuations. Note that  $T_c$  is in the range between the melting temperature  $T_m$  and the glass temperature  $T_g$ , e.g.  $T_m > T_c > T_g$ .

It is really the  $\alpha$ -process exists also below  $T_c$ . This process leads to a very slow decay of the apparently frozen structures, i.e. the nonergodic structures obtained from the MCT [1,4] are approximately stable only for a finite time interval. This slow decay shows the typical properties which correspond usually to the dynamics of the main glass transition (WLF like behavior of the relaxation time, stretched exponential decay of the correlation function). This effects can be partially described in terms of an extended mode coupling theory [9,10] introducing additional hopping processes. There exists also various alternative descriptions [3,11] which explain the cooperative motion of the particles inside a supercooled liquid below  $T_c$ . One of these possibilities is the  $n$ -spin facilitated kinetic Ising model [11–14], originally introduced by Fredrickson and Andersen. The base idea of this model consists in a coarse graining of space and time scales and simultaneously a reduction of the degrees of freedom. In detail that means:

1. *Coarse graining of spatial scales:* The supercooled liquid is separated into cells, so that each cell contains a sufficiently large number of particles which realize a representative number of molecular motions, i.e. the many body system is considered of a virtual lattice with the unit size  $l$ . This lattice has no influence on the underlying dynamics of the supercooled liquid.
2. *Reduction of the degrees of freedom:* Each cell will be characterized by only one trivial degree of freedom, i.e. the cell structure enables us to attach to each cell an observable  $\sigma_j$  which characterizes the actual state of particles inside the cell  $j$ . The usual realization is given by the local density  $\rho_j$  (particles per cell) with  $\sigma_j = -1$  if  $\rho_j > \bar{\rho}$  and  $\sigma_j = 1$  if  $\rho_j < \bar{\rho}$  where  $\bar{\rho}$  is the averaged density of the system. This mapping implies consequently different mobilities of the particles inside such a cell, i.e.  $\sigma_j = -1$  corresponds to a more immobile (or solid like) state and  $\sigma_j = 1$  to a more mobile (or liquid like) state of the cell  $j$ . The set of all observables  $\sigma = \{\sigma_j\}$  forms a configuration. The evolution of the statistical probability distribution function  $P(\sigma, t)$  can be described by a generalized master equation [15] using a projection of the real dynamics onto the dynamics of  $\sigma$ :

$$\frac{\partial P(\sigma, t)}{\partial t} = \sum_{\sigma'} L'(\sigma, \sigma') P(\sigma, t) + \sum_{\sigma'} \int_0^t K(\sigma, \sigma', t - t') P(\sigma, t') dt'$$

3. *Coarse graining of the time scale:* The last step bases on the assumption that possible memory terms  $K(\sigma, \sigma', t)$  of the generalized master equation is mainly determined by fast molecular processes while the slow dynamics is mainly reflected by the temporally local contributions  $\sum_{\sigma'} L'(\sigma, \sigma') P(\sigma, t)$ . Of course, the validity of this assumption depends strongly on the choice of the remaining degrees of freedom, and in many cases it is very hard (or impossible from the actual point of view) to give a satisfactory explanation of this assumption. However, if this separation of the dynamics is justified, an elementary time scale larger than the time scale of the fast molecular processes can be introduced. Therefore, the memory will be reduced to simple temporally local terms, i.e.  $K(\sigma, \sigma', t - t') = \delta(t - t') \int_0^\infty K(\sigma, \sigma', \tau) d\tau$ . One obtains an evolution equation which is equivalent to the mathematical representation of a usual master equation.

$$\frac{\partial P(\sigma, t)}{\partial t} = \sum_{\sigma'} L(\sigma, \sigma') P(\sigma, t) \quad (1)$$

The dynamical matrix  $L(\sigma, \sigma') = L'(\sigma, \sigma') + \int_0^\infty K(\sigma, \sigma', \tau) d\tau$  is determined by the above discussed formal procedure. Unfortunately, a direct calculation is mostly very complicated, so that one should use reasonable assumptions about the mathematical structure of  $L$ .

To make the time evolution of the glass configurations more transparent we use the argumentation following the idea of Fredrickson and Andersen [11–14], i.e. we suppose that the basic dynamics is a simple (Glauber) process  $\sigma_j = +1 \leftrightarrow \sigma_j = -1$  controlled by the thermodynamical Gibbs measure and by self induced topological restrictions. In particular, an elementary flip at a given cell is allowed only if the number of the nearest neighbored mobile cells ( $\sigma_j = +1$ ) is equal or larger than a restriction number  $n$  with  $0 < n < z_c$  ( $z_c$ : coordination number of the lattice). Elementary flip processes and geometrical restrictions lead to the cooperative rearrangement of the underlying system and therefore to mesoscopical models describing a supercooled liquid below  $T_c$ . These models [11–14] are denoted as  $n$ -spin facilitated Ising model on a  $d$ -dimensional lattice SFM[ $n, d$ ]. The self-adapting environments influence in particular the long time behavior of the spin-spin correlation functions and therefore of the corresponding density-density correlation functions. The SFM[ $n, d$ ] was studied numerically [16–19] (SFM[2, 2]) and recently also analytically [20] (SFM[1, 1]).

From this point of view it will be an interesting task to derive a set of equations related to the SFM[ $n, d$ ] which are similar to the well known Mori–Zwanzig equations [21] and which can be used as a reasonable basis for a further treatment, e.g. for a continuous fraction analysis or for a mode coupling approach.

The aim of the present paper is the derivation of such evolution equations and their analysis in terms of a mode coupling approach. We restrict our investigation to the analysis of the SFM[2,  $d$ ] but a generalization to another class of spin facilitated kinetic Ising models is always possible. The starting point is the mapping of the master equations of the SFM[2,  $d$ ] to evolution equations in a Fock-space representation. Using a projection formalism one obtains evolution equations for a set of relevant observables and consequently for the corresponding correlation functions. The paper ends in an analysis of these correlation functions in terms of the frequency matrices and memory terms.

## II. FOCK-SPACE APPROACH

Following Doi [22], compare also [23], the probability distribution  $P(\sigma, t)$  can be related to a state vector  $|F(t)\rangle$  in a Fock-space according to  $P(\sigma, t) = \langle \sigma | F(t) \rangle$  and  $|F(t)\rangle = \sum_{\sigma} P(\sigma, t) |\sigma\rangle$ , respectively, with the base vectors  $|\sigma\rangle$ . Using this representation, the Master equation (1) can be transformed to an equivalent equation in a Fock-space

$$\partial_t |F(t)\rangle = \hat{L} |F(t)\rangle \quad (2)$$

The dynamical matrix  $L(\sigma, \sigma')$  of (1) is now mapped onto the operator  $\hat{L}$  given in a second quantized form with  $d$  and  $d^\dagger$  being the annihilation and creation operators, respectively, for flips processes. Usually  $\hat{L}$  is expressed in terms of creation and annihilation operators which satisfy Bose commutation rules [22, 24, 25].

The SFM[ $n, d$ ] can be interpreted as a lattice gas ( $\sigma_i = 0$ : empty cell,  $\sigma_i = 1$ : occupied cell) considering the excluded volume effect, i.e. changes of the configuration  $\sigma$  are possible only under the presence of the exclusion principle. To

preserve the restriction of the occupation number in the underlying dynamical equations too, the commutation rules of the operators  $\hat{d}$  and  $\hat{d}^\dagger$  are chosen as those of Pauli-operators [23,24,26,27]:

$$[\hat{d}_i, \hat{d}_j^\dagger] = \delta_{i,j}(1 - 2\hat{d}_i^\dagger \hat{d}_i) \quad [\hat{d}_i, \hat{d}_j] = [\hat{d}_i^\dagger, \hat{d}_j^\dagger] = 0 \quad \hat{d}_i^2 = (\hat{d}_i^\dagger)^2 = 0 \quad (3)$$

It should be remarked that the method can be extended to the case of higher restricted occupation numbers [29]. As it was shown by Doi [22] the average of a physical quantity  $B(\sigma)$  is given by the average of the corresponding operator  $\hat{B}(t) = \sum_\sigma |\sigma\rangle B(\sigma) \langle\sigma|$  via

$$\langle \hat{B}(t) \rangle = \sum_\sigma P(\sigma, t) B(\sigma) = \langle s | \hat{B} | F(t) \rangle \quad (4)$$

using the reference state  $\langle s | = \sum_\sigma \langle \sigma |$ . The normalization condition is manifested in the relation  $\langle s | F(t) \rangle = 1$ . In the same way, arbitrary correlation functions can be expressed by

$$\langle \hat{A}(t) \hat{B}(t') \rangle = \sum_{\sigma, \sigma'} A(\sigma) P(\sigma, t; \sigma', t') B(\sigma') = \langle s | \hat{A} \exp \{ \hat{L}(t - t') \} \hat{B} | F(t') \rangle$$

From this point of view, it is possible to create the evolution equations of various averages and correlation functions. For example, using (2) and (4) one obtains the evolution equation for an arbitrary operator  $\hat{B}$  [28]:

$$\partial_t \langle \hat{B} \rangle = \langle s | [\hat{B}, \hat{L}] | F(t) \rangle \quad (5)$$

Here we have used the necessary relation  $\langle s | \hat{L} = 0$ , which is an immediately consequence of the normalization condition. The evolution operator for the SFM[2,  $d$ ] can be written as

$$\hat{L} = \sum_{i,j,k} \kappa_{i|jk} \hat{D}_j \hat{D}_k \left[ \beta(\hat{d}_i - \hat{D}_i) + \lambda(\hat{d}_i^\dagger - (1 - \hat{D}_i)) \right] \quad (6)$$

with the particle number operator  $\hat{D}_i = \hat{d}_i^\dagger \hat{d}_i$  and temperature dependent jumping rates  $\lambda$  and  $\beta$ .  $\kappa_{i|jk}$  is a lattice function with  $\kappa_{i|jk} = 1$  if  $j \neq k$  and  $j$  and  $k$  are neighbored to lattice cell  $i$ . Applying a simple activation dynamics one obtains for the jumping rates:

$$\beta = \nu^{-1}(T) \quad \text{and} \quad \lambda = \nu^{-1}(T) \exp(-\varepsilon/T) \quad (7)$$

where  $\nu^{-1}(T)$  is an elementary temperature dependent time scale ( $\varepsilon$  is the energy difference between the solid and liquid like state). Note that the stationary state corresponds to an average  $\bar{\sigma}_{\text{eq}} = \langle \hat{D}_j \rangle = \lambda/(\lambda + \beta)$  which can be obtained directly from (5). The knowledge of  $\hat{L}$  and the corresponding evolution equation (2) allows a reasonable analysis of the SFM[2,  $d$ ].

The Fock space formalism has the decisive advantage of a simple construction principle for each evolution operator  $\hat{L}$  on the basis of creation and annihilation operators. Therefore, this method allows investigations of master equations for various evolution processes, e.g. aggregation, chemical reactions [29,30], nonlinear diffusion [31] and just the spin facilitated kinetic Ising model.

### III. PROJECTION EQUATIONS

#### A. relevant operators

The dynamics of an arbitrary physical system can be described by a reasonable set of relevant operators. We use the normalized local deviations of the configuration from the thermodynamical average and the corresponding derivatives with respect to the time as suitable relevant observables for the investigation of the SFM[2,  $d$ ]

$$\hat{\eta}_i^{(0)}(t) = \hat{\eta}_i(t) = \frac{\hat{D}_i(t) - \bar{\sigma}_{\text{eq}}}{\sqrt{\bar{\sigma}_{\text{eq}}(1 - \bar{\sigma}_{\text{eq}})}} \quad \text{and} \quad \hat{\eta}_i^{(\beta)}(t) = \frac{\partial^\beta \hat{\eta}_i(t)}{\partial t^\beta} = \hat{\eta}_i(t) \hat{L}^\beta \quad (8)$$

( $\beta = 0, 1, \dots, g_{\max}$ ; usually the upper borderline is a finite integer number, but  $g_{\max} \rightarrow \infty$  is also possible). These covariant operators must be extended by the corresponding contravariant operators

$$\tilde{\eta}_i^{(0)}(t) = \hat{\eta}_i(t) \quad \text{and} \quad \tilde{\eta}_i^{(\beta)}(t) = \hat{L}^\beta \hat{\eta}_i(t) \quad (9)$$

Using (8) and (9) we construct the backward projection operator  $\hat{P}$ :

$$\hat{P} = \sum_{\alpha, \beta, i, j} \left\langle \dots \tilde{\eta}_i^{(\alpha)} \right\rangle g_{ij}^{\alpha\beta} \tilde{\eta}_j^{(\beta)} \quad \text{with} \quad \sum_{\alpha, i} \left\langle \hat{\eta}_k^{(\gamma)} \tilde{\eta}_i^{(\alpha)} \right\rangle g_{ij}^{\alpha\beta} = \delta^{\gamma\beta} \delta_{kj} \quad (10)$$

(with  $\alpha, \beta, \dots \in [0, g_{\max}]$ ). The projection operator leads to an identical mapping of the relevant operators onto itself, i.e.  $\hat{\eta}_k^{(\gamma)} \hat{P} = \hat{\eta}_k^{(\gamma)}$ . Consequently, the orthogonal projection operator  $\hat{Q}$  is given by  $\hat{Q} = 1 - \hat{P}$  with  $\hat{\eta}_k^{(\gamma)} \hat{Q} = 0$ .

## B. Basis equations

The evolution equation (2) leads to the formal solution  $|F(t)\rangle = \exp\{\hat{L}t\} |F(0)\rangle$ . The dependence of  $|F(t)\rangle$  on the time can be transferred to an arbitrary operator analogous to the transformation of Schrödinger's representation into the Heisenberg picture. Therefore one obtains time-dependent operators

$$\hat{B}(t) = \hat{B} \exp\{\hat{L}t\} \quad (11)$$

The derivation of the evolution equations for the relevant observables starts from the formal time evolution of  $\hat{\eta}_k^{(\gamma)}(t)$ . It follows from (11)

$$\frac{\partial \hat{\eta}_k^{(\gamma)}(t)}{\partial t} = \hat{\eta}_k^{(\gamma)}(t) \hat{L} \quad (12)$$

This equation is the basis for the derivation of projection equations for the relevant observables in analogy to the well known Mori-Zwanzig [21] equations for classical or quantum mechanical equations of motion. The application of  $1 = \hat{P} + \hat{Q}$  onto the operator  $\hat{L}$  leads to a formal splitting into a relevant and an irrelevant part (Note that  $\hat{P}$  realizes a projection onto the subspace  $L_{\parallel}$  of relevant operators, whereas  $\hat{Q}$  projects onto the linearly independent subspace  $L_{\perp}$  of all other operators). Hence,

$$\begin{aligned} \frac{\partial \hat{\eta}_k^{(\gamma)}(t)}{\partial t} &= \hat{\eta}_k^{(\gamma)} \exp\{\hat{L}t\} \hat{L} = \hat{\eta}_k^{(\gamma)} \hat{L} (\hat{P} + \hat{Q}) \exp\{\hat{L}t\} \\ &= - \sum_{\beta, j} \Omega_{kj}^{(\gamma\beta)} \hat{\eta}_j^{(\beta)} + \hat{\eta}_k^{(\gamma)} \hat{L} \hat{Q} \exp\{\hat{L}t\} \end{aligned} \quad (13)$$

with the frequency matrix

$$\Omega_{kj}^{(\gamma\beta)} = - \sum_{\alpha, i} \left\langle \hat{\eta}_k^{(\gamma)} \hat{L} \tilde{\eta}_i^{(\alpha)} \right\rangle g_{ij}^{\alpha\beta} \quad (14)$$

The second term of (13) can be rewritten by using an identical transformation of  $\exp\{\hat{L}t\}$  into an integral expression:

$$\exp\{\hat{L}t\} = \int_0^t dt' \exp\{\hat{L}\hat{Q}(t-t')\} \hat{L} \hat{P} \exp\{\hat{L}t'\} + \exp\{\hat{L}\hat{Q}t\} \quad (15)$$

This relation allows the derivation of rigorous projection equations similar to the usual Mori-Zwanzig-equations [21,32]:

$$\frac{\partial \hat{\eta}_k^{(\gamma)}(t)}{\partial t} = - \sum_{\beta, j} \Omega_{kj}^{(\gamma\beta)} \hat{\eta}_j^{(\beta)} + \int_0^t dt' \sum_{\beta, j} K_{kj}^{(\gamma\beta)}(t-t') \hat{\eta}_j^{(\beta)}(t') + \hat{f}_k^{(\gamma)}(t) \quad (16)$$

with the residual forces

$$\hat{f}_k^{(\gamma)}(t) = \hat{\eta}_k^{(\gamma)}(t) \hat{L} \hat{Q} \exp \left\{ \hat{L} \hat{Q} t \right\} = \hat{f}_k^{(\gamma)} \exp \left\{ \hat{L} \hat{Q} t \right\} \quad (17)$$

(with the properties  $\hat{f}_k^{(\gamma)}(t) \hat{Q} = \hat{f}_k^{(\gamma)}(t)$  and  $\hat{f}_k^{(\gamma)}(t) \hat{P} = 0$ ) and the memory matrix:

$$K_{kj}^{(\gamma\beta)}(t-t') = \sum_{\alpha,i} \left\langle \hat{\eta}_k^{(\gamma)} \hat{L} \hat{Q} \exp \left\{ \hat{L} \hat{Q} (t-t') \right\} \hat{L} \hat{\eta}_i^{(\alpha)} \right\rangle g_{ij}^{\alpha\beta} \quad (18)$$

The comparison between (16) and the usual Mori-Zwanzig-equations shows a formal equivalence. Both equations contains frequency terms, memory and residual forces with a similar mathematical structure. But there is a fundamental difference which can be studied directly by inspecting the memory kernel. The memory of Mori-Zwanzig-equations can be written always as a correlation function of the residual forces. This relation can be interpreted as a representation of the fluctuation-dissipation theorem, and it is causally connected with the fact, that the Mori-Zwanzig equations are related to reversible classical or quantum mechanical equations. On the other hand, the memory (18) cannot be completely constructed from residual forces (17). The cause is the irreversible character of the underlying master equation.

### C. Projection equations for a reduced set of relevant observables

We choose  $g_{\max} = 1$  for the following investigations, i.e. the relevant observables are  $\hat{\eta}_i^{(0)}(t) = \hat{\eta}_i(t)$  and  $\hat{\eta}_i^{(1)}(t) = \hat{\eta}_i(t) \hat{L}$ . This settling corresponds slightly to mechanical systems, which are completely determined by spatial coordinates and velocities. The general system of equations (16) becomes

$$\begin{aligned} \frac{\partial}{\partial t} \hat{\eta}_n^{(0)}(t) &= \sum_j \sum_{\beta=0,1} \left[ -\Omega_{nj}^{(0\beta)} \hat{\eta}_j^{(\beta)}(t) + \int_0^t dt' K_{nj}^{(0\beta)}(t-t') \hat{\eta}_j^{(\beta)}(t') \right] + \hat{f}_n^{(0)}(t) \\ \frac{\partial}{\partial t} \hat{\eta}_n^{(1)}(t) &= \sum_j \sum_{\beta=0,1} \left[ -\Omega_{nj}^{(1\beta)} \hat{\eta}_j^{(\beta)}(t) + \int_0^t dt' K_{nj}^{(1\beta)}(t-t') \hat{\eta}_j^{(\beta)}(t') \right] + \hat{f}_n^{(1)}(t) \end{aligned} \quad (19)$$

A simple analysis leads to the simplifications  $\Omega_{nj}^{(0\beta)} = -\delta_{nj} \delta^{1\beta}$ ,  $\hat{f}_n^{(0)}(t) = 0$  and  $K_{nj}^{(0\beta)}(t-t') = 0$ . Therefore, the first equation will be reduced to the identity  $\partial_t \hat{\eta}_n^{(0)}(t) = \partial_t \hat{\eta}_n(t) = \hat{\eta}_n^{(1)}(t)$  and the second equation can be rewritten as

$$\begin{aligned} \frac{\partial^2}{\partial t^2} \hat{\eta}_n(t) &= -\sum_j \left[ \Omega_{nj}^{(10)} \hat{\eta}_j(t) + \Omega_{nj}^{(11)} \frac{\partial}{\partial t} \hat{\eta}_j(t) \right] \\ &+ \sum_j \int_0^t dt' \left[ K_{nj}^{(10)}(t-t') \hat{\eta}_j(t') + K_{nj}^{(11)}(t-t') \frac{\partial}{\partial t'} \hat{\eta}_j(t') \right] + \hat{f}_n^{(1)}(t) \end{aligned} \quad (20)$$

The result is a second order differential equation which reflects the complete dynamics of the relevant observables.

### D. Projection equations for correlation functions

An important rule for experimental and theoretical investigations of the glass transition plays the time-dependent equilibrium correlation functions of the relevant observables. Especially the SFM $[n, d]$  should be characterized by

$$\Phi_{nm}(t) = \langle \hat{\eta}_n(t) \hat{\eta}_m(0) \rangle = \left\langle s \left| \hat{\eta}_n \exp \left\{ \hat{L} t \right\} \hat{\eta}_m \right| F(0) \right\rangle \quad (21)$$

These correlation function is equivalent to the normalized spin-spin correlation:

$$\Phi_{nm}(t) = \frac{\langle \sigma_n(t) \sigma_m(0) \rangle - \bar{\sigma}_{\text{eq}}^2}{\bar{\sigma}_{\text{eq}}(1 - \bar{\sigma}_{\text{eq}})}$$

which should be similar to the normalized density-density correlation of the underlying supercooled liquid, i.e.  $\Phi_{nm}(t) \sim \langle \delta \rho(\mathbf{r}, t) \delta \rho(\mathbf{r}', 0) \rangle$ , see also the above discussed mapping  $\rho \rightarrow \sigma$ . The evolution equation of  $\Phi_{nm}(t)$  follows from (20) by a right hand multiplication with  $\hat{\eta}_m$  and a subsequent determination of the average. The contributions of the residual forces  $\hat{f}_n^{(1)}(t)$  vanish identically. One obtains a homogeneous integro-differential equation:

$$\begin{aligned} \frac{\partial^2 \Phi_{nm}}{\partial t^2} = & - \sum_j \left[ \Omega_{nj}^{(10)} \Phi_{jm} + \Omega_{nj}^{(11)} \frac{\partial \Phi_{jm}}{\partial t} \right] \\ & + \int_0^t dt' \left[ K_{nj}^{(10)}(t-t') \Phi_{jm}(t') + K_{nj}^{(11)}(t-t') \frac{\partial \Phi_{jm}(t')}{\partial t} \right] \end{aligned} \quad (22)$$

This evolution equation of  $\Phi_{nm}(t)$  is a rigorous second order integro-differential equation, which will be analyzed now. To this aim it is necessary to determine the frequency and memory parts for the thermodynamical equilibrium. Here, correlation functions, memory and frequency matrices should be homogeneous and isotropic functions, e.g.  $\Phi_{mn}(t) = \Phi(|\mathbf{n} - \mathbf{m}|, t)$  or  $K_{mn}^{(\alpha\beta)}(t) = K^{(\alpha\beta)}(|\mathbf{n} - \mathbf{m}|, t)$ . The homogeneity is a direct consequence of the underlying translation invariance. On the other hand, isotropy can be expected for the asymptotic case of the continuous limit, i.e. for  $|\mathbf{n} - \mathbf{m}| \rightarrow \infty$ . However, the isotropy is partially disturbed at finite distances as a consequence of the underlying lattice structure. The Fourier transformation of (22) can be obtained by using the representation

$$\Phi_{mn}(t) = \sum_{\mathbf{q}} \Phi(\mathbf{q}, t) \exp \{i\mathbf{q}(\mathbf{n} - \mathbf{m})\} \iff \Phi(\mathbf{q}, t) = \frac{1}{N} \sum_n \Phi(|\mathbf{n}|, t) \exp \{-i\mathbf{q}\mathbf{n}\}$$

of the correlation function whereas the frequency matrix and the memory can be written as:

$$\Omega^{(il)}(\mathbf{q}) = \frac{1}{N} \sum_n \Omega^{(il)}(|\mathbf{n}|) \exp \{-i\mathbf{q}\mathbf{n}\} \quad (23)$$

and

$$K^{(il)}(\mathbf{q}, t-t') = \frac{1}{N} \sum_n K^{(il)}(|\mathbf{n}|, t-t') \exp \{-i\mathbf{q}\mathbf{n}\} \quad (24)$$

respectively.  $\mathbf{n}$  and  $\mathbf{m}$  denote the lattice vector of size  $n$  and  $m$ ,  $\mathbf{q}$  is a vector of the first Brillouin zone corresponding to the lattice. We use a cubic lattice for the following calculations but an application of another lattice type is always possible. The Mori-Zwanzig-equation (22) becomes now:

$$\begin{aligned} \ddot{\Phi}(q, t) = & -N \left[ \Omega^{(10)}(q) \Phi(q, t) + \Omega_{nj}^{(11)}(q) \dot{\Phi}(q, t) \right] \\ & + N \int_0^t dt' \left[ K^{(10)}(q, t-t') \Phi(q, t') + K^{(11)}(q, t-t') \dot{\Phi}(q, t') \right] \end{aligned} \quad (25)$$

Note that all quantities depend only on  $q = |\mathbf{q}|$  (at least for the continuous limit) because of the isotropy. Finally, the Laplace transformation

$$\Phi(q, z) = \int_0^\infty dt \exp \{-zt\} \Phi(q, t) \quad (26)$$

leads to the algebraic equation

$$\Phi(q, z) = \frac{\Phi_0(q)}{z + \frac{N\Omega^{(10)}(q) - NK^{(10)}(q, z) - zg_0(q)}{z + N\Omega^{(11)}(q) - NK^{(11)}(q, z) + g_0(q)}} \quad (27)$$

which considers the initial conditions  $\Phi(q, 0) = \Phi_0(q)$  and  $\dot{\Phi}(q, 0) = \dot{\Phi}_0(q)$ . Furthermore, the quantity  $g_0(q)$  denotes the ratio  $g_0(q) = \dot{\Phi}_0(q)/\Phi_0(q)$ . It should be remarked that especially  $\Omega^{(11)}(q) \neq 0$  and  $K^{(10)}(q, z) \neq 0$  are consequences of the irreversible master equations. On the other hand, the usual Mori-Zwanzig equations are founded on reversible Liouville operators which lead immediately to  $\Omega^{(11)}(q) = 0$  and  $K^{(10)}(q, z) = 0$ .

#### IV. DETERMINATION OF FREQUENCY MATRICES

The concrete determination of the frequency matrices is possible by using the concrete evolution operator  $\hat{L}$  (6). Note that the projection equations (22) are valid for an arbitrary physical system which can be described by master equations.

The frequency matrices contain always the matrix  $\mathbf{g}$ , which can be calculated from (10), i.e.  $\mathbf{g}$  is determined by the following system of linear equations:

$$h_{ik}^{(\alpha\gamma)} = \left\langle \hat{\eta}_i^{(\alpha)} \tilde{\eta}_k^{(\gamma)} \right\rangle \quad \text{with} \quad \sum_{\gamma,k} h_{ik}^{(\alpha\gamma)} g_{kj}^{\gamma\beta} = \delta^{\alpha\beta} \delta_{ij} \quad (28)$$

Using the definition

$$\Gamma_{ik}^\beta = \left\langle \hat{\eta}_i \hat{L}^\beta \hat{\eta}_k \right\rangle \quad (29)$$

one obtains simple expressions for the matrix  $h$  (i.e.  $h_{ik}^{(\alpha\gamma)} = \Gamma_{ik}^{\alpha+\gamma}$ ) as well as for the frequency matrix (14):

$$\Omega_{kj}^{(\gamma\beta)} = - \sum_{\alpha,i} \Gamma_{ki}^{\alpha+\gamma+1} g_{ij}^{\alpha\beta} \quad (30)$$

The knowledge of  $\Gamma_{ik}^\beta$  ( $\beta = 0 \dots 3$ ) allows the exact determination of  $\Omega^{(10)}(q)$  and  $\Omega_{nj}^{(11)}(q)$ . The values  $\Gamma_{ik}^\beta$  can be obtained straightforwardly by using (6) and the commutation relations (3). It follows:

$$\Gamma_{mn}^\alpha = \left( \frac{1}{-\tau_0} \right)^\alpha (A^\alpha \delta_{mn} + B^\alpha \Theta_{nm} + C^\alpha \chi_{mn} + D^\alpha \zeta_{nm}) \quad (31)$$

The lattice functions  $\Theta_{nm}$ ,  $\chi_{mn}$  and  $\zeta_{nm}$  vanish, except for the following cases:  $\Theta_{nm} = 1$  for  $|\mathbf{m} - \mathbf{n}| = 1$ ,  $\chi_{mn} = 1$  for  $|\mathbf{m} - \mathbf{n}| = \sqrt{2}$  and  $\zeta_{nm} = 1$  for  $|\mathbf{m} - \mathbf{n}| = 2$ . The values  $A^\alpha$ ,  $B^\alpha$ ,  $C^\alpha$  and  $D^\alpha$  are listed in appendix A. The Fourier transformation is now a simple calculation. The approximation for small wave vectors (continuous limit) is by a special interest. The actual lattice structure becomes irrelevant on these sufficiently large spatial scales, i.e. the Fourier transformed  $\Gamma_{mn}^\alpha$  are isotropic values. One obtains up to the second order in  $q$ :

$$\Gamma^\alpha(q) = \frac{1}{N} \frac{1}{(-\tau_0)^\alpha} (\Gamma_0^\alpha - \gamma^\alpha q^2) \quad (32)$$

The coefficients  $\Gamma_0^\alpha$  and  $\gamma^\alpha$  follows immediately from the values  $A^\alpha$ ,  $B^\alpha$ ,  $C^\alpha$  and  $D^\alpha$ :

$$\Gamma_0^\alpha = A^\alpha + z_c B^\alpha + \frac{1}{2} z_c (z_c - 2) C^\alpha + z_c D^\alpha \quad \text{and} \quad \gamma^\alpha = B^\alpha + (z_c - 2) C^\alpha + 4 D^\alpha \quad (33)$$

( $z_c$  is the coordination number of the  $d$ -dimensional lattice, i.e.  $z_c$  is the number of nearest neighbors per lattice cell. Straightforwardly, the Fourier transformed matrix  $g^{\alpha\beta}(q)$  can be written as

$$g^{(\alpha\beta)}(q) = \frac{1}{WN} \begin{pmatrix} \Gamma_0^2 - \gamma^2 q^2 & \tau_0 \Gamma_0^1 \\ \tau_0 \Gamma_0^1 & \tau_0^2 \end{pmatrix} \quad (34)$$

with  $W = \Gamma_0^2 - (\Gamma_0^1)^2 - \gamma^2 q^2$ . Finally, the frequency matrices  $\Omega^{(10)}(q)$  and  $\Omega^{(11)}(q)$  are given by

$$N\Omega^{(10)}(q) = \frac{1}{W\tau_0^2} \left\{ \Gamma_0^1 \Gamma_0^3 - (\Gamma_0^2)^2 + (2\Gamma_0^2 \gamma^2 - \Gamma_0^1 \gamma^3) q^2 \right\} \quad (35)$$

and

$$N\Omega^{(11)}(q) = \frac{1}{W\tau_0} \left\{ \Gamma_0^3 - \Gamma_0^1 \Gamma_0^2 + (\Gamma_0^1 \gamma^2 - \gamma^3) q^2 \right\} \quad (36)$$

in the continuous limit. Note, that (34), (35) and (36) considers already that  $\gamma^0 = \gamma^1 = 0$  and  $\Gamma_0^0 = 1$ .

## V. ANALYSIS OF THE RELAXATION BEHAVIOR

A rough understanding of the results so far is possible by an analysis of the relaxation behavior of the correlation function  $\Phi(q, t)$  neglecting the memory terms containing in (27). In this case the correlation function is reduced to a finite continued fraction. It can be expected that this case is related to the high temperature limit corresponding to a more or less exponential decay of the correlation function. Furthermore, this approximation should be reasonable for the description of the correlation function at short time scales. Note that because of  $K^{(10)}(q, t) \rightarrow \text{const.}$  and  $K^{(11)}(q, t) \rightarrow \text{const.}$  for  $t \rightarrow 0$ , one obtains  $K^{(10)}(q, z) \sim z^{-1}$  and  $K^{(11)}(q, z) \sim z^{-1}$  for  $z \rightarrow \infty$ . Therefore, the

memory terms can be neglected at sufficiently short time scales  $t \rightarrow 0$  or  $z \rightarrow \infty$ . The initial conditions  $\Phi_0(q)$  and  $\dot{\Phi}_0(q)$  of the correlation function  $\Phi$  are defined by equilibrium averages:

$$\Phi_{nm}(0) = \langle \hat{\eta}_n \hat{\eta}_m \rangle = \Gamma_{nm}^0 \quad \text{and} \quad \dot{\Phi}_{nm}(0) = \langle \hat{\eta}_m \hat{L} \hat{\eta}_n \rangle = \Gamma_{nm}^1 \quad (37)$$

and consequently

$$g_0(q) = \frac{\dot{\Phi}_0(q)}{\Phi_0(q)} = N\Gamma^1(q) \quad (38)$$

The normalized correlation function  $\tilde{\Phi}(q, z) = \Phi(q, z)/\Phi_0(q)$  follows from (27) under consideration of (38) and under neglect of memory terms. A simple calculation leads to

$$\tilde{\Phi}(q, z) = \frac{z + N\Omega^{(11)}(q) + N\Gamma^1(q)}{z^2 + zN\Omega^{(11)}(q) + N\Omega^{(10)}(q)} \quad (39)$$

which can be written as

$$\tilde{\Phi}(q, z) = \frac{A_1}{z - z_1} + \frac{A_2}{z - z_2} \quad (40)$$

with the poles

$$z_{1/2} = -\frac{1}{2} \left[ N\Omega^{(11)}(q) \mp \sqrt{(N\Omega^{(11)}(q))^2 - 4N\Omega^{(10)}(q)} \right] \quad (41)$$

and the intensities

$$A_1 = \frac{N\Gamma^1(q) - z_2}{z_1 - z_2} \quad A_2 = \frac{z_1 - N\Gamma^1(q)}{z_1 - z_2}$$

The present approximation of the SFM[2,  $d$ ] is characterized by two relaxation times  $\tau_R^1(q) = z_1^{-1}$  and  $\tau_R^2(q) = z_2^{-1}$ . It can be verified by a simple calculation that both relaxation times shows only a weak dependence on  $q$ . Furthermore, both relaxation times approach finite values for  $q \rightarrow 0$ . Obviously, the spin facilitated kinetic Ising model shows no diffusion-like modes which behave as  $\tau \sim q^{-2}$  for the limit  $q \rightarrow 0$ . This finding agrees with investigations of the one dimensional spin facilitated kinetic Ising model [33]. The SFM[1, 1] corresponds to diffusion processes combined with creation and annihilation processes of active states. A cell has an active state if this cell can change its state without any support by further flip processes of neighbored cells. Creation and annihilation processes dominates at sufficiently large scales, i.e. an inhomogeneity reaching over a sufficiently large distance will be reduced by local creation and annihilation processes of mobile cells until diffusion processes becomes effective.

As above mentioned, we restrict our investigations to the borderline case of macroscopic scales, i.e.  $q \rightarrow 0$ . Fig.1 shows the relaxation times  $\tau_R^1(q)$  and  $\tau_R^2(q)$  as a function of temperature. As expected, there is no significant difference between the relaxation times for  $q = 0$  and  $q \neq 0$ , respectively. Furthermore, the slow relaxation time  $\tau_R^1(q)$  shows a weak non-Arrhenius behavior. One obtains  $\ln \tau_R^2 \sim \ln \nu(T) + o(q^2)$  (see also eq.7) and  $\ln \tau_R^1 \sim \ln \nu(T) + 2\varepsilon/T + o(q^2)$ , respectively, at low temperatures. On the other hand, the high temperature regime is characterized by another temperature dependence:  $\ln \tau_R^{1,2} = \ln \nu(T) + u^{1,2}\varepsilon/T + o(q^2)$ . The coefficients  $u^{1,2}$  depend on the actual lattice structure and the spatial dimension. But a simple analysis shows that always  $u^1 < 2$ , i.e. the activation energy of the SFM[2,  $d$ ] increases with decreasing temperature.

The existence of two relaxation times means not that the SFM[2,  $d$ ] is characterized by an  $\alpha$ - and a  $\beta$ -process. The superposition of both decays,  $\exp\{-t/\tau_R^1\}$  and  $\exp\{-t/\tau_R^2\}$ , considering of the intensities  $A_1$  and  $A_2$  (see fig.2), shows a continuous decay of the correlation function  $\tilde{\Phi}(q, t)$ , see fig.3. This behavior is in an agreement with numerical simulations [19,16] and it corresponds also to the above discussed thought, that spin facilitated kinetic Ising models are possible candidates modelling the behavior of supercooled liquids below the critical temperature of the usual mode coupling theory.

## VI. DETERMINATION OF THE MEMORY MATRICES

## A. Complete and orthogonal basis

All operators acting on the Fock-space can be represented by a complete collection of orthogonal base operators. The determination of such a basis is possible under consideration of the underlying  $\hat{d}_i, \hat{d}_i^\dagger$ - (pseudo fermionic) algebra (3) of the SFM[2,  $d$ ]. The base operators can be expressed as all possible products of the above introduced operators  $\hat{\eta}_i$  (8). A base operator is denoted as  $\hat{B}_{\mathbf{N}_n}^{(n)}$ . (The index  $n$  corresponds to the order of the product,  $\mathbf{N}_n$  is an  $n$ -dimensional vector indicating the concerning lattice cells). The first groups of the basis are:

$$\begin{aligned}\hat{B}^{(0)} &= 1 \\ \hat{B}_i^{(1)} &= \hat{\eta}_i \\ \hat{B}_{ij}^{(2)} &= \hat{\eta}_i \hat{\eta}_j \quad \text{for } i < j \\ \hat{B}_{ijk}^{(3)} &= \hat{\eta}_i \hat{\eta}_j \hat{\eta}_k \quad \text{for } i < j < k\end{aligned}$$

Note that because of the commutation relation  $[\hat{\eta}_i, \hat{\eta}_j] = 0$  the components of  $\mathbf{N}_n$  can be ordered. The case of two or more equivalent indices is excluded because  $\hat{\eta}_i^2 = (1 - 2\bar{\sigma}_{\text{eq}})/\bar{\sigma}_{\text{eq}} \hat{\eta}_i + 1$ , i.e. quadratic or higher powers of each operator  $\hat{\eta}_i$  can be always reduced to a linear representation. The orthogonality means that:

$$\langle \hat{B}_{\mathbf{N}_n}^{(n)} \hat{B}_{\mathbf{N}_m}^{(m)} \rangle = \delta^{nm} \delta_{\mathbf{N}_n, \mathbf{N}_m} \quad (42)$$

This relation can be checked considering that all equilibrium averages of operators on various cells decay in a product of averages with respect to these cells, e.g.  $\langle \hat{\eta}_i^2 \dots \hat{\eta}_j \dots \hat{\eta}_k^2 \dots \rangle = \langle \hat{\eta}_i^2 \rangle \dots \langle \hat{\eta}_j \rangle \dots \langle \hat{\eta}_k^2 \rangle \dots$ . This important relation is valid for all SFM[ $n, d$ ] if the neighbor-neighbor interaction vanishes. Note that the Hamiltonian of the analyzed class of spin facilitated kinetic Ising models is given by  $\hat{H} = \sum_i \varepsilon \hat{D}_i(t) \simeq \sum_i \varepsilon \sqrt{\bar{\sigma}_{\text{eq}}} (1 - \bar{\sigma}_{\text{eq}}) \hat{\eta}_i(t)$ . From this point of view, the relation (42) follows immediately because of  $\langle \hat{\eta}_i \rangle = 0$  and  $\langle \hat{\eta}_i^2 \rangle = 1$ . Thus, the basis  $\tilde{B} = \{ \hat{B}_{\mathbf{N}_n}^{(n)} \}$  is orthogonal.

The completeness of  $\tilde{B} = \{ \hat{B}_{\mathbf{N}_n}^{(n)} \}$  is to be understood in relation to the reference state  $\langle s |$ , i.e. the following equation is fulfilled for an arbitrary operator  $\hat{X}$ :

$$\langle s | \hat{X} = \sum_n \sum_{\mathbf{N}_n} \langle \hat{X} \hat{B}_{\mathbf{N}_n}^{(n)} \rangle \langle s | \hat{B}_{\mathbf{N}_n}^{(n)} \quad (43)$$

The mathematical proof of this property is given in appendix B.

## B. Decomposition of the memory terms

Eq.18 is a reasonable starting point for an analysis of the memory terms. The consideration of (8) and (9) leads to:

$$K_{kj}^{(\gamma\beta)}(t) = \sum_{\alpha, i} \langle \hat{\eta}_k \hat{L}^{\gamma+1} \hat{Q} \exp \{ \hat{Q} \hat{L} \hat{Q} t \} \hat{Q} \hat{L}^{\alpha+1} \eta_i \rangle g_{ij}^{\alpha\beta} \quad (44)$$

(Note that  $\hat{Q}^2 = \hat{Q}$ ). The application of the completeness relation (43) onto (44) yields:

$$K_{kj}^{(\gamma\beta)}(t) = \sum_{\alpha, i} \sum_{n, m} \sum_{\mathbf{N}_n, \mathbf{N}_m} H_{k, \mathbf{N}_n}^{\gamma(n)} \langle \hat{B}_{\mathbf{N}_n}^{(n)} \exp \{ \hat{Q} \hat{L} \hat{Q} t \} \hat{B}_{\mathbf{N}_m}^{(m)} \rangle \tilde{H}_{\mathbf{N}_m, i}^{(m)\alpha} g_{ij}^{\alpha\beta} \quad (45)$$

with

$$H_{k, \mathbf{N}_n}^{\gamma(n)} = \langle \hat{\eta}_k \hat{L}^{\gamma+1} \hat{Q} \hat{B}_{\mathbf{N}_n}^{(n)} \rangle \quad \text{and} \quad \tilde{H}_{\mathbf{N}_n, k}^{(n)\gamma} = \langle \hat{B}_{\mathbf{N}_n}^{(n)} \hat{Q} \hat{L}^{\gamma+1} \eta_k \rangle \quad (46)$$

These coefficients can be determined by simple algebraic calculations. One obtains immediately that both,  $H_{k, \mathbf{N}_n}^{\gamma(n)}$  and  $\tilde{H}_{\mathbf{N}_n, i}^{(n)\alpha}$  vanish identically for  $n = 0, 1$ . On the other hand, the coefficients (46) vanish also identically for  $n > 5$  because  $\gamma \leq 1$ . Hence, the memory terms can be constructed by using a finite number of functions  $\langle \hat{B}_{\mathbf{N}_n}^{(n)} \exp \{ \hat{Q} \hat{L} \hat{Q} t \} \hat{B}_{\mathbf{N}_m}^{(m)} \rangle$ . These functions will be transformed identically. One obtains:

$$\begin{aligned}
\langle \hat{B}_{\mathbf{N}_n}^{(n)} \exp \{ \hat{Q} \hat{L} \hat{Q} t \} \hat{B}_{\mathbf{N}_m}^{(m)} \rangle &= \langle \hat{B}_{\mathbf{N}_n}^{(n)} \exp \{ \hat{Q} \hat{L} \hat{Q} t \} \exp \{ -\hat{L} t \} \exp \{ \hat{L} t \} \hat{B}_{\mathbf{N}_m}^{(m)} \rangle \\
&= \sum_p \sum_{\mathbf{N}_p} \langle \hat{B}_{\mathbf{N}_n}^{(n)} \exp \{ \hat{Q} \hat{L} \hat{Q} t \} \exp \{ -\hat{L} t \} \hat{B}_{\mathbf{N}_p}^{(p)} \rangle \langle \hat{B}_{\mathbf{N}_p}^{(p)} \exp \{ \hat{L} t \} \hat{B}_{\mathbf{N}_m}^{(m)} \rangle \\
&= \sum_p \sum_{\mathbf{N}_p} \langle \hat{B}_{\mathbf{N}_n}^{(n)} \exp \{ \hat{Q} \hat{L} \hat{Q} t \} \exp \{ -\hat{L} t \} \hat{B}_{\mathbf{N}_p}^{(p)} \rangle \langle \hat{B}_{\mathbf{N}_p}^{(p)}(t) \hat{B}_{\mathbf{N}_m}^{(m)} \rangle
\end{aligned} \tag{47}$$

The averages  $\langle \hat{B}_{\mathbf{N}_p}^{(p)}(t) \hat{B}_{\mathbf{N}_m}^{(m)} \rangle$  are usual many point correlation functions.

### C. Mode coupling approximation

#### 1. Short time evolution of the memory

An exact determination of  $\langle \hat{B}_{\mathbf{N}_n}^{(n)} \exp \{ \hat{Q} \hat{L} \hat{Q} t \} \exp \{ -\hat{L} t \} \hat{B}_{\mathbf{N}_p}^{(p)} \rangle$  and  $\langle \hat{B}_{\mathbf{N}_p}^{(p)}(t) \hat{B}_{\mathbf{N}_m}^{(m)} \rangle$  may be possible only for some few special cases. Therefore, we need a suitable approximation for a further treatment. In a first step we analyze the function:

$$\Psi_{\mathbf{N}_n \mathbf{N}_p}^{(np)}(t) = \langle \hat{B}_{\mathbf{N}_n}^{(n)} \exp \{ \hat{Q} \hat{L} \hat{Q} t \} \exp \{ -\hat{L} t \} \hat{B}_{\mathbf{N}_p}^{(p)} \rangle \tag{48}$$

(47) can be interpreted as a separation of fast and slow time scales. The operator  $\hat{Q} \hat{L} \hat{Q}$  is related to a dynamics completely different to the dynamics of  $\hat{L}$ . In general, it can be expected that  $\hat{B}_{\mathbf{N}_n}^{(n)} \exp \{ \hat{Q} \hat{L} \hat{Q} t \}$  shows a significant evolution on a very short time scale in comparison to the characteristic time scale related to  $\hat{B}_{\mathbf{N}_n}^{(n)} \exp \{ \hat{L} t \}$ . Therefore, we come to the rough conclusion: while the evolution operator  $\hat{L}$  contains all relevant time scales, the operator  $\hat{Q} \hat{L} \hat{Q}$  is mainly determined by contributions related to short time scales, i.e. the long time contributions are partially cancelled by the projection procedure. Consequently, the term  $\exp \{ \hat{Q} \hat{L} \hat{Q} t \} \exp \{ -\hat{L} t \}$  and therefore  $\Psi_{\mathbf{N}_n \mathbf{N}_p}^{(np)}(t)$  should be dominated by long time scales, because the fast time scales are eliminated at least partially by the factor  $\exp \{ -\hat{L} t \}$ . Clearly, this is a very rough interpretation, but it gives an explanation for the assumption that the time dependence of  $\Psi_{\mathbf{N}_n \mathbf{N}_p}^{(np)}(t)$  is weak in comparison to the decay of the correlation function  $\langle \hat{B}_{\mathbf{N}_p}^{(p)}(t) \hat{B}_{\mathbf{N}_m}^{(m)} \rangle$  which is connected only with the time evolution factor  $\exp \{ \hat{L} t \}$ .

However, it seems to be reasonable to expand  $\Psi_{\mathbf{N}_n \mathbf{N}_p}^{(np)}(t)$  in powers of the time  $t$ . The determination of all Taylor coefficients is out of the question. But an approximative analysis is possible by using a finite number of coefficients. We hope that the error of this perturbation expansion is sufficiently strong suppressed by the corresponding factor  $\langle \hat{B}_{\mathbf{N}_p}^{(p)}(t) \hat{B}_{\mathbf{N}_m}^{(m)} \rangle$  (see eq.47). One obtains:

$$\Psi_{\mathbf{N}_n \mathbf{N}_p}^{(np)}(t) = \sum_{M=0}^{\infty} \Lambda_{\mathbf{N}_n, \mathbf{N}_p}^{(np), M} \frac{t^M}{M!}$$

with:

$$\Lambda_{\mathbf{N}_n, \mathbf{N}_p}^{(np), M} = \left\langle \hat{B}_{\mathbf{N}_n}^{(n)} \frac{\partial^M}{\partial t^M} \left[ \exp \{ \hat{Q} \hat{L} \hat{Q} t \} \exp \{ -\hat{L} t \} \right]_{t=0} \hat{B}_{\mathbf{N}_p}^{(p)} \right\rangle$$

The first coefficients  $\Lambda_{\mathbf{N}_n, \mathbf{N}_p}^{(np), M}$  can be determined by simple calculations, e.g.

$$\begin{aligned}
\Lambda_{\mathbf{N}_n, \mathbf{N}_p}^{(np), 0} &= \langle \hat{B}_{\mathbf{N}_n}^{(n)} \hat{B}_{\mathbf{N}_p}^{(p)} \rangle = \delta^{np} \delta_{\mathbf{N}_n, \mathbf{N}_p} \\
\Lambda_{\mathbf{N}_n, \mathbf{N}_p}^{(np), 1} &= \langle \hat{B}_{\mathbf{N}_n}^{(n)} (\hat{Q} \hat{L} \hat{Q} - \hat{L}) \hat{B}_{\mathbf{N}_p}^{(p)} \rangle
\end{aligned} \tag{49}$$

In principle, the discussed expansion is an exact representation (The radius of convergence of the exponential function is infinite large). The true approximation consists in the breaking of the Taylor expansion after a finite power of  $t$ . We restrict our investigation to the simplest case, i.e. we assume  $\Lambda_{\mathbf{N}_n, \mathbf{N}_p}^{(np), M} = 0$  for  $M \geq 1$ . Hence, one obtains

$$\Psi_{\mathbf{N}_n \mathbf{N}_p}^{(np)}(t) \approx \delta^{np} \delta_{\mathbf{N}_n, \mathbf{N}_p} \quad (50)$$

But it should be remarked that an extension to higher terms is possible without any problems. We abstain from a consideration of higher terms with respect to the clarity of the calculations. Furthermore, the obtained results (see below) using (50) show already a reasonable agreement with numerical simulations.

## 2. Decomposition of the many point correlation functions

The main problem is a reasonable approximation of the function  $\langle \hat{B}_{\mathbf{N}_p}^{(p)}(t) \hat{B}_{\mathbf{N}_m}^{(m)} \rangle$ . This function decays in products of simple pair correlation functions if the distances between the corresponding lattice points (defined by the vectors  $\mathbf{N}_p$  and  $\mathbf{N}_m$ ) are sufficiently large:

$$\langle \hat{B}_{(i_1 i_2 \dots i_p)}^{(p)}(t) \hat{B}_{(j_1 j_2 \dots j_p)}^{(m)} \rangle \simeq \frac{1}{p!} (\Phi_{i_1 j_1}(t) \Phi_{i_2 j_2}(t) \dots \Phi_{i_p j_p}(t) + \text{perm}) \quad (51)$$

This asymptotic limit is correct for infinitely large (or at least sufficiently large) distances between the lattice cells  $i_1, i_2, \dots$ . We use this borderline case as an approximation for an arbitrary set of lattice cells  $\{\mathbf{N}_p, \mathbf{N}_m\}$ . This approximation is equivalent to the decomposition of higher static correlation functions into simple pair correlation functions. For example, a similar approach was used to create self consistent equations for the static structure factor [34]. Furthermore, this approximation is also the kernel of the well known mode coupling approach [35].

## 3. Reduction of the basis

The third approximation consists in a reduction of the basis  $\tilde{B} = \{\hat{B}_{\mathbf{N}_n}^{(n)}\}$ . It was demonstrated that only base operators with  $n \leq 5$  are necessary for a representation of (46). Because of the approximations (50) and (51) all other factors of (45) contains also no higher base operators  $\hat{B}_{\mathbf{N}_n}^{(n)}$ . The consideration of all relevant  $\hat{B}_{\mathbf{N}_n}^{(n)}$  is no general problem. However, we restrict the calculations only to elements with  $n \leq 2$ , also with respect to clarity. Really, the neglected terms containing base operators  $\hat{B}_{\mathbf{N}_n}^{(n)}$  with  $5 \geq n \geq 3$  yield only small additional contributions to the final results. Thus, the complete representation of the memory (45) is given by the approximation:

$$K_{kj}^{(\gamma\beta)}(t) \approx \frac{1}{2} \sum_{\alpha, i} \sum_{i_1 i_2 j_1 j_2} H_{k, (i_1 i_2)}^{\gamma(2)} [\Phi_{i_1 j_1}(t) \Phi_{i_2 j_2}(t) + \Phi_{i_1 j_2}(t) \Phi_{i_2 j_1}(t)] \tilde{H}_{(j_1 j_2).i}^{(2)\alpha} g_{ij}^{\alpha\beta} \quad (52)$$

## D. Macroscopic scale

As above mentioned, the  $q$ -dependence of the frequency matrices is very small. This weak dependence can be expected also for the memory terms, i.e. we restrict our investigations only to the macroscopic scale  $q \rightarrow 0$ . The assumption of a weak dependence on  $q$  leads to:

$$\Phi_{nm}(t) = \varphi(t) \delta_{nm} = \frac{1}{N} \sum_{\mathbf{q}} \varphi(t) \exp \{i \mathbf{q}(\mathbf{n} - \mathbf{m})\} \quad (53)$$

This approximation is related to the fact that all correlation functions between different lattice cells  $\Phi_{nm}(t)$  with  $n \neq m$  (see eq.21) vanish for  $t = 0$  ( $\Phi_{nm}(0)$  is the equilibrium average  $\langle \hat{\eta}_n \hat{\eta}_m \rangle$  which decouples as a result of the simple Hamiltonian  $\hat{H} \simeq \sum_i \hat{\eta}_i(t)$ , i.e.  $\langle \hat{\eta}_n \hat{\eta}_m \rangle = \langle \hat{\eta}_n \rangle \langle \hat{\eta}_m \rangle = 0$ ) and for  $t \rightarrow \infty$  (because of the ergodicity follows again  $\Phi_{nm}(\infty) = \langle \hat{\eta}_n(\infty) \hat{\eta}_m(0) \rangle = \langle \hat{\eta}_n(\infty) \rangle \langle \hat{\eta}_m(0) \rangle = 0$ ). Furthermore, one obtains only a very small correlation  $\Phi_{nm}(t)$  between different cells for finite times  $t$  differences which can be checked by numerical investigation. From (38) and (32) it follows for  $q \rightarrow 0$  the relation  $g_0(0) = -\Gamma_0^1/\tau_0$ . Thus, the evolution equation on a macroscopic scale is given by (Note, that there is the initial condition:  $\varphi(0) = 1$ ):

$$\varphi(z) = \left[ z + \frac{N\Omega^{(10)}(0) - NK^{(10)}(0, z) + z\Gamma_0^1/\tau_0}{z + N\Omega^{(11)}(0) - NK^{(11)}(0, z) - \Gamma_0^1/\tau_0} \right]^{-1} \quad (54)$$

The determination of the frequency matrices was realized above. One obtains in the macroscopic limit by using (35) and (36):

$$N\Omega^{(10)}(0) = \frac{1}{\tau_0^2} \frac{\Gamma_0^1 \Gamma_0^3 - (\Gamma_0^2)^2}{\Gamma_0^2 - (\Gamma_0^1)^2} \quad \text{and} \quad N\Omega^{(11)}(0) = \frac{1}{\tau_0} \frac{\Gamma_0^3 - \Gamma_0^1 \Gamma_0^2}{\Gamma_0^2 - (\Gamma_0^1)^2} \quad (55)$$

It remains the determination of the memory. Using the definition:

$$\begin{aligned} h^\alpha(\mathbf{q}, \mathbf{q}') &= \frac{1}{N} \sum_{j,k,l} H_{j,(kl)}^{\alpha(2)} \exp\{i\mathbf{q}(\mathbf{k} - \mathbf{j}) + i\mathbf{q}'(\mathbf{l} - \mathbf{j})\} \\ \tilde{h}^\alpha(\mathbf{q}, \mathbf{q}') &= \frac{1}{N} \sum_{j,k,l} \tilde{H}_{(kl),j}^{(2)\alpha} \exp\{i\mathbf{q}(\mathbf{k} - \mathbf{j}) + i\mathbf{q}'(\mathbf{l} - \mathbf{j})\} \end{aligned}$$

and (53), it follows from (52):

$$\begin{aligned} NK^{(\gamma\beta)}(0, t) &= \frac{1}{N} \sum_{\alpha} \sum_{\mathbf{q}} h^\gamma(\mathbf{q}, -\mathbf{q}) \varphi(t)^2 \tilde{h}^\alpha(-\mathbf{q}, \mathbf{q}) (Ng^{\alpha\beta}(0)) \\ &\approx \sum_{\alpha} h^\gamma(0, 0) \varphi(t)^2 \tilde{h}^\alpha(0, 0) Ng^{\alpha\beta}(0) \end{aligned}$$

The substitution  $h^\gamma(\mathbf{q}, -\mathbf{q}) \rightarrow h^\gamma(0, 0)$  and  $\tilde{h}^\alpha(-\mathbf{q}, \mathbf{q}) \rightarrow \tilde{h}^\alpha(0, 0)$  is possible because the  $q$ -dependence of these quantities is again relatively weak. We need  $h^1(0, 0)$ ,  $\tilde{h}^0(0, 0)$  and  $\tilde{h}^1(0, 0)$  for the following investigations. A simple calculation leads to  $\tilde{h}^0(0, 0) = 0$ . Using (34) the memory terms can be written as:

$$\begin{aligned} NK^{(10)}(0, t) &= \tau_0 \varphi(t)^2 h^1(0, 0) \tilde{h}^1(0, 0) \frac{\Gamma_0^1}{\Gamma_0^2 - (\Gamma_0^1)^2} \\ NK^{(11)}(0, t) &= \tau_0^2 \varphi(t)^2 h^1(0, 0) \tilde{h}^1(0, 0) \frac{1}{\Gamma_0^2 - (\Gamma_0^1)^2} \end{aligned}$$

It should be remarked that the ratio between both memory terms is given by the relation:

$$\frac{K^{(10)}(0, t)}{K^{(11)}(0, t)} = \frac{\Gamma_0^1}{\tau_0}$$

Finally, we must determine the quantity  $\lambda = \tau_0^2 h^1(0, 0) \tilde{h}^1(0, 0) / (4(\Gamma_0^2 - (\Gamma_0^1)^2))$ . This is again an algebraic procedure which can be realized straightforwardly. The final results are very unwieldy. Therefore, we give only the explicit expressions for the asymptotic case of low temperatures, i.e. for  $\bar{\sigma}_{\text{eq}} \rightarrow 0$ . One obtains for a square lattice ( $z_c = 4$ ) and a cubic lattice ( $z_c = 6$ ), respectively:

$$\begin{aligned} \lambda &= \frac{32\bar{\sigma}_{\text{eq}}^3}{3} (1 + o(\bar{\sigma}_{\text{eq}})) \quad \text{for} \quad z_c = 4 \\ \lambda &= \frac{64\bar{\sigma}_{\text{eq}}^3}{5} (1 + o(\bar{\sigma}_{\text{eq}})) \quad \text{for} \quad z_c = 6 \end{aligned}$$

The behavior  $\lambda \sim \bar{\sigma}_{\text{eq}}^3$  is characteristic for  $T \rightarrow 0$  and  $\bar{\sigma}_{\text{eq}} \rightarrow 0$ , respectively. (54) can now be written as:

$$\varphi(z) = \left[ z + \frac{\Gamma_0^1}{\tau_0} - \frac{1}{\tau_0} \frac{\Gamma_0^1 N\Omega^{(11)}(0)\tau_0 - N\Omega^{(10)}(0)\tau_0^2 - (\Gamma_0^1)^2}{z\tau_0 + N\Omega^{(11)}(0)\tau_0 - \lambda\Xi(z) - \Gamma_0^1} \right]^{-1} \quad (56)$$

with  $\Xi(z) = \int_0^\infty (dt/\tau_0) \varphi(t)^2 \exp\{-zt\}$ .

## VII. DISCUSSION

Now we are able to analyze the characteristic slowing down of the dynamics of the SFM[2,  $d$ ] for decreasing temperature. The central equation for the following discussion is (56). The first question is the existence of ergodicity and nonergodicity. Exists there a critical temperature  $T^*$ , so that the correlation function  $\varphi(t)$  shows an incomplete decay  $\varphi(t \rightarrow \infty) = f_\infty \neq 0$  for  $T \leq T^*$ ? In other words, has the function  $\varphi(z)$  a pole at  $z = 0$  below the critical temperature  $T^*$ ? This question is equivalent to the determination of a kinetic phase transition from an ergodic state into a nonergodic state for supercooled liquids [1, 4]. To this aim we split the correlation function into a nonergodicity part  $f_\infty$  and a contribution  $\varphi_{\text{erg}}(t)$ :

$$\varphi(t) = f_\infty + \varphi_{\text{erg}}(t) \quad (57)$$

The function  $\varphi_{\text{erg}}(t)$  describes the remaining ergodic part of the SFM[2,  $d$ ], i.e.  $\varphi_{\text{erg}}(t \rightarrow \infty) = 0$ . The Laplace transformation leads to

$$\varphi(z) = \frac{f_\infty}{z} + \varphi_{\text{erg}}(z) \quad (58)$$

with  $\lim_{z \rightarrow 0} z\varphi_{\text{erg}}(z) = 0$ . The memory term  $\Xi(z)$  can be expressed by:

$$\Xi(z) = \frac{f_\infty^2}{z\tau_0} + \Xi_{\text{erg}}(z) \quad (59)$$

with  $\lim_{z \rightarrow 0} z\Xi_{\text{erg}}(z) = 0$ . (58), (59) and (56) yield:

$$\lim_{z \rightarrow 0} z\varphi(z) = f_\infty = \lim_{z \rightarrow 0} \left[ 1 + \frac{\Gamma_0^1}{z\tau_0} + \frac{\Gamma_0^1 N \Omega^{(11)}(0) \tau_0 - N \Omega^{(10)}(0) \tau_0^2 - (\Gamma_0^1)^2}{\lambda f_\infty^2} \right]^{-1} \quad (60)$$

It follows immediately that the nonergodicity part  $f_\infty$  has a nonvanishing value only if  $\Gamma_0^1 = 0$ . Otherwise, the only solution of (60) is given by  $f_\infty = 0$ , i.e. the SFM[2,  $d$ ] is an ergodic system if  $\Gamma_0^1 \neq 0$ . The value of  $\Gamma_0^1$  vanishes only for  $T = 0$ :

$$\Gamma_0^1 = 0 \iff T = 0$$

i.e. the SFM[2,  $d$ ] realizes a kinetic phase transition from an ergodic system to a nonergodic system at the critical temperature  $T^* = 0$ . Thus, the only nonergodic state can be observed at zero-point temperature. Additionally, we obtain from (55):

$$\Omega^{(11)}(0) = 0 \quad \text{and} \quad \Omega^{(10)}(0) = 0 \quad \text{for} \quad T = 0$$

Thus, the nonergodic part is given by:

$$f_\infty = 1$$

i.e. an initial equilibrium configuration at  $T = 0$  shows no structure relaxations during the total observation time. This behavior is a consequence of the fact that the kinetic phase transition occurs at the absolute zero at temperature. In other words, each arbitrary equilibrium configuration is frozen at  $T = T^* = 0$ .

We obtain the important result that the SFM[2,  $d$ ] is ergodic for all finite temperatures  $T > 0$ . This is a real contradiction to the statements of the original papers [11,12] which predict a kinetic phase transition at a finite critical temperature.

Of course, it is possible to explain  $T^* = 0$  by a simple picture. A finite fraction of active cells (i.e. cells which are able to change their state without any previous change of states of neighbored cells) exists at each temperature  $T > 0$ . The concentration of these active or nonfrozen cells is proportional to  $\bar{\sigma}_{\text{eq}}^2$  ( $\bar{\sigma}_{\text{eq}}^2$  is equivalent to the probability that a cell has two neighbored cells with  $\sigma = 1$ ). Active cells are mainly isolated at sufficiently low temperatures, but an annihilation of an isolated active cell is never possible. The property to be an active cell can be transferred to a neighbored cell by some few elementary flip processes, (this procedure can be interpreted as a diffusion of active states), a new active cell can be created in the nearest environment of an initially active cell (creation process) and two neighbored active cells can be unified to one active region and further this region can be reduced to one active cell (annihilation process). Diffusion, annihilation and creation are well-balanced in the thermodynamical equilibrium. On the other hand, this three processes realize a motion of active states through the whole volume, i.e. each cell is able to change their state after a sufficiently long waiting time. Note that this considerations must be modified for a finite volume because some configurations of the SFM[2,  $d$ ] show a self blockade in finite geometries [19]. But it should be denoted also, that a special choice of the initial configuration excludes any type of self blockades [16]. However, the main result of our discussion is the ergodicity of the SFM[2,  $d$ ] for  $T > 0$ .

To analyze the relaxation behavior at finite temperatures  $T > 0$  we introduce the relaxation time  $\tau_c$

$$\tau_c = \tau_0 \frac{N \Omega^{(11)}(0) \tau_0 - \lambda \Xi(0) - \Gamma_0^1}{N \Omega^{(10)}(0) \tau_0^2 - \lambda \Gamma_0^1 \Xi(0)} \quad (61)$$

and the coefficient

$$\varsigma = N\Omega^{(11)}(0)\tau_0 - \lambda\Xi(0) - \Gamma_0^1 \quad (62)$$

Using these notations, we get from (56):

$$\varphi(z) = \left[ z + \tau_c^{-1} + \left( \frac{\Gamma_0^1}{\tau_0} - \tau_c^{-1} \right) \left[ 1 + \frac{\varsigma}{z\tau_0 + \lambda[\Xi(0) - \Xi(z)]} \right]^{-1} \right]^{-1} \quad (63)$$

(61), (62) and (63) are a closed, nonlinear system of equations, which can be solved by numerical standard methods. Fig.4 shows  $\varphi(t)$  for a SFM[2, 3] on a square lattice ( $z_c = 6$ ) for various relative temperatures  $T/\varepsilon$ . We see that the correlation function  $\varphi(t)$  shows with decreasing temperature a pronounced stretched decay over some decades, while an exponential-like decay is obtained for high temperatures. This stretching can be illustrated by a simple argument. Short times ( $t \rightarrow 0$  or  $z \rightarrow \infty$ ) are related to a behavior  $\varphi(z) \simeq (z + \Gamma_0^1/\tau_0)^{-1}$  or  $\varphi(t) \simeq \exp\{-\Gamma_0^1 t/\tau_0\}$ . On the other hand, the long time regime ( $t \rightarrow \infty$  or  $z \rightarrow 0$ ) is characterized by  $\varphi(z) \simeq (z + \tau_c^{-1})^{-1}$  or  $\varphi(t) \simeq \exp\{-t/\tau_c\}$ . As there is  $\Gamma_0^1/\tau_0 \gg \tau_c^{-1}$  we expect a typical crossover between both regimes characterized by a stretched decay, see fig.4. But it should be remarked, that the decay of  $\varphi(t)$  is no Kohlrausch-Williams-Watts function ( $\simeq \exp(-(t/\tau)^\gamma)$ ). There exists only over a finite interval a reasonable fit with such a stretched exponential function [37].

The spectral density  $S(\lambda_s)$  of the correlation function is defined as the set of amplitudes of exponential decays which contribute to  $\varphi(t)$ . Thus,  $\varphi(t)$  is the Laplace transformed spectral density with respect to the Laplace variable  $t$ :

$$\varphi(t) = \int_0^\infty d\lambda_s S(\lambda_s) \exp(-\lambda_s t) \quad (64)$$

One obtains the remarkable result that the spectral density is positive definite, (fig.5). The knowledge of the spectral densities allows the determination of other interesting properties, for example the susceptibility  $\chi(\omega)$ , see fig. 6.

Finally, the averaged relaxation time  $\tau(T)$  can be obtained by using

$$\tau(T) = \frac{\int_0^\infty t \varphi(t) dt}{\int_0^\infty \varphi(t) dt} = \frac{\int_0^\infty S(\lambda_s) \lambda_s^{-2} d\lambda_s}{\int_0^\infty S(\lambda_s) \lambda_s^{-1} d\lambda_s} \quad (65)$$

This relaxation time shows with decreasing temperature an increasing deviation from the simple relaxation times (41). Especially, one obtains now a typical non-Arrhenius behavior (fig.7).

## VIII. CONCLUSIONS

It was shown that irreversible master equations can be easily transformed into projection equations by using the Fock space representation. Whereas the usual projection formalisms, which leads to the well known Mori-Zwanzig equations, start from a reversible Liouville equation, the master equations are already irreversible. As a consequence of this initial irreversibility, one obtains additional frequency matrices and memory terms. Thus, these additional contributions are caused mainly by the loss of the invariance against an inversion of the time.

A second important property follows from a general analysis of the frequency matrices. The corresponding poles of the Laplace transformed correlation function  $\hat{\Phi}(q, z)$  are always located on the negative real axis. Especially, the imaginary part of the poles vanishes identically. This behavior is related to the general structure of the master equation. The dynamical matrix  $L(\sigma, \sigma')$  of a master equation is always negative definite (or better semidefinite because at least one eigen value is zero as result of the conservation of the probability). Thus, only relaxation processes should be observed, i.e. the evolution of the probability  $P(\sigma, t)$  can be approached by a probably infinitely large expansion in terms of exponential functions:  $P(\sigma, t) = P_{\text{eq}}(\sigma, t) + \sum_m A_m(\sigma) \exp\{-\Lambda_m t\}$  with ( $\Lambda_m > 0$  for all  $m$ ). One obtains no oscillations in contradiction to microscopical systems which bases on a Liouville equation. The absent of oscillations is directly connected with the Markov property of the underlying master equation. This behavior corresponds also to the fact that the observation of a spin wave propagation (corresponding to density waves in a glass or a supercooled liquid) is not possible for the SFM[ $n, d$ ]. From this point of view, the traditional notation 'frequency matrix' can be misleading. It seems to be possibly favorable to use the notion relaxation matrix. However, we have used the

traditional terminology to avoid misunderstandings and conflicts with other well defined quantities.

In principle, the consideration of all possible time derivations  $\hat{\eta}_i^{(\beta)}(t) = \partial^\beta \hat{\eta}_i(t) / \partial t^\beta = \hat{\eta}_i(t) \hat{L}^\beta$  ( $\beta = 0, 1, \dots, \infty$ ) as relevant operators leads to an infinite continuous fraction for the correlation function  $\Phi_{nm}(t)$ , determined by frequency matrices of various order. This representation allows a systematic analysis of the short time behavior of the SFM[2,  $d$ ], because an infinite continuous fraction contains no memory term. Unfortunately, a general explicit determination of the frequency matrices, e.g. by a successive rule, cannot be obtained. On the other hand, each approximation using a finite number of frequency matrices (e.g.  $\Omega^{(10)}$  and  $\Omega^{(11)}$  in the present case) fails for sufficiently long times. However, some important properties of the spin facilitated kinetic Ising model can be verified for this relatively rough approximation, e.g. a weak non-Arrhenius behavior of the relaxation times. Note, that this result is in agreement with various numerical simulations [16,19]. On the other hand, the typical stretched decay of the correlation function can not be explained by using this simple approximation.

A satisfactory treatment is possible by an approximative consideration of the memory terms. This procedure leads to an equation, which is partially similar to the well known mode coupling equation of supercooled liquids. The memory terms yield the main contributions to the typical stretching of the correlation function. Some small remaining deviations from the numerical results are caused by the approximations (50) and (51) of the memory terms.

From this point of view, we come back to the initial question. Which processes of the usual glass transition can be described by the spin facilitated kinetic Ising models? Obviously, the spin facilitated kinetic Ising model is not adequate for a description of the fast processes inside a supercooled liquid. This statement is supported by both, numerical simulations [19] and the presented analytical investigations, which show that no fast ( $\beta$ -) processes can be observed. On the other hand, spin facilitated kinetic Ising models allow a more or less reasonable, quantitative description of the slow ( $\alpha$ -) process in supercooled liquids below the critical temperature  $T_c$  of the usual mode coupling theory [1,4]. Note that for this low temperature regime the time scales of  $\alpha$ - and  $\beta$ -process are well separated, i.e. a separation of the underlying dynamic is actually possible and it is considered in the structure of the master equations related to the SFM[ $n, d$ ], see above. The fast dynamics, corresponding to the  $\beta$ -process, determines the thermodynamical noise. This noise is not explicitly contained in the previous equations, but it is the underlying cause for the irreversibility of the master equations. Additionally, master equations and stochastic evolution equations are principally equivalent [36]. However, the remaining slow dynamics of a supercooled liquid ( $\alpha$ -process) is represented in the kinetic scenario of the SFM[ $n, d$ ].

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## APPENDIX A: REPRESENTATION OF $\Gamma$

The rigorous values  $A^\alpha$ ,  $B^\alpha$ ,  $C^\alpha$  and  $D^\alpha$  of (33) can be obtained straightforwardly by using the evolution operator  $\hat{L}$  (6), the commutation relations (3) and the definition of  $\Gamma_{ik}^\beta$  (29). Under consideration of the coordination number  $z_c$  and the thermodynamical equilibrium  $\bar{\sigma}_{\text{eq}}$  of the cell state we obtain the following results  $A^\alpha$ -terms:

$$\begin{aligned} A^0 &= 1 \\ A^1 &= 2\bar{\sigma}_{\text{eq}}^2 \binom{z_c}{2} \\ A^2 &= 4\bar{\sigma}_{\text{eq}}^2 \left[ \binom{z_c}{2} + 6\bar{\sigma}_{\text{eq}} \binom{z_c}{3} + 6\bar{\sigma}_{\text{eq}}^2 \binom{z_c}{4} \right] \\ A^3 &= 8\bar{\sigma}_{\text{eq}}^2 \left[ \binom{z_c}{2} + 24\bar{\sigma}_{\text{eq}} \binom{z_c}{3} + 114\bar{\sigma}_{\text{eq}}^2 \binom{z_c}{4} + 180\bar{\sigma}_{\text{eq}}^3 \binom{z_c}{5} + 90\bar{\sigma}_{\text{eq}}^4 \binom{z_c}{6} \right] \\ &\quad + 4\bar{\sigma}_{\text{eq}}^3 (1 - \bar{\sigma}_{\text{eq}}) z_c (z_c - 1)^2 (1 + \bar{\sigma}_{\text{eq}}(z_c - 2)) (2 + \bar{\sigma}_{\text{eq}}(z_c - 4)) \end{aligned}$$

$B^\alpha$ -terms:

$$\begin{aligned} B^0 &= 0 \\ B^1 &= 0 \\ B^2 &= 4\bar{\sigma}_{\text{eq}}^3 (1 - \bar{\sigma}_{\text{eq}})(z_c - 1)^2 \\ B^3 &= 16\bar{\sigma}_{\text{eq}}^3 (1 - \bar{\sigma}_{\text{eq}}) z_c (z_c - 1)^2 (1 + 3\bar{\sigma}_{\text{eq}}(z_c - 2) + \bar{\sigma}_{\text{eq}}^2(z_c - 2)(z_c - 3)) \end{aligned}$$

$C^\alpha$ -terms:

$$C^0 = C^1 = C^2 = 0$$

and

$$C^3 = 8\bar{\sigma}_{\text{eq}}^3 (1 - \bar{\sigma}_{\text{eq}})^2 (2 + 2\bar{\sigma}_{\text{eq}} z_c (z_c - 2))$$

$D^\alpha$ -terms:

$$D^0 = D^1 = D^2 = 0$$

and

$$D^3 = 8\bar{\sigma}_{\text{eq}}^4 (1 - \bar{\sigma}_{\text{eq}})^2 (z_c - 1)^2$$

## APPENDIX B: COMPLETENESS OF THE BASIS $\tilde{B}$

The proof consists in two parts. At first we analyze an operator  $\hat{X}'$ , which is a multilinear form of the operators  $\hat{\eta}_i$ , i.e.

$$\hat{X}' = \sum_n \sum_{\mathbf{N}_n} \beta_{\mathbf{N}_n}^{(n)} \hat{B}_{\mathbf{N}_n}^{(n)} \quad (\text{B1})$$

with arbitrary coefficients  $\beta_{\mathbf{N}_n}^{(n)}$ . The orthogonality of the basis  $\tilde{B} = \{\hat{B}_{\mathbf{N}_n}^{(n)}\}$  leads immediately to  $\beta_{\mathbf{N}_n}^{(n)} = \langle \hat{X}' \hat{B}_{\mathbf{N}_n}^{(n)} \rangle$ , i.e. all coefficients of  $\hat{X}'$  can be determined by a successive procedure. In other words, the basis  $\{\hat{B}_{\mathbf{N}_n}^{(n)}\}$  is complete with respect all operators  $\hat{X}'$ , i.e. the basis forms a space containing all operators of type (B1). The second part of the proof analyses arbitrary operators  $\hat{X}$  acting on the Fock-space.  $\hat{X}$  consists in operators  $\hat{\eta}_k$  as well as annihilation operators  $\hat{d}_k$  and creation operators  $\hat{d}_k^\dagger$ . A representation like (B1), extended by the operators  $\hat{d}_k$  and  $\hat{d}_k^\dagger$  is always possible:

$$\hat{X} = \sum_{\mathbf{M}^1, \mathbf{M}^2, \mathbf{M}^3, \mathbf{M}^4} \theta_{\mathbf{M}^1, \mathbf{M}^2, \mathbf{M}^3, \mathbf{M}^4} \prod_{k=1}^N \left[ \hat{\eta}_k^{m_k^1} \hat{d}_i^{m_k^2} \left( \hat{d}_i^\dagger \right)^{m_k^3} \hat{1}^{m_k^4} \right] \quad (\text{B2})$$

( $\hat{1}$  is the simple unit operator  $\hat{1} \equiv 1$ ,  $N$  is the number of lattice cells). The vectors  $\mathbf{M}^\gamma$  ( $\gamma = 1, \dots, 4$ ) contains  $N$  integer numbers, i.e.  $\mathbf{M}^\gamma = \{m_1^\gamma, \dots, m_N^\gamma\}$  with  $m_k^\gamma \geq 0$  for all possible  $k$  and  $\gamma$ . The commutation relations (3) can be used to write  $\hat{X}$  as a representation with the internal restrictions:  $m_k^1 + m_k^2 + m_k^3 + m_k^4 = 1$  for all lattice cells  $k$ , i.e. each contribution to the sum (B2) contains exactly one of the four operators  $\hat{1}$ ,  $\hat{d}_k$ ,  $\hat{d}_k^\dagger$  and  $\hat{\eta}_k$  with respect to any cell  $k$ . Furthermore, the commutation relation (3) allows a shift of all operators related to a given lattice cell  $i$  to the left hand side. It is simple to show that [28]:

$$\begin{aligned} \langle s | 1 &= \langle s | 1 & \langle s | \hat{d}_i &= \langle s | \left( 1 - \bar{\sigma}_{\text{eq}} - \hat{\eta}_i \sqrt{\bar{\sigma}_{\text{eq}}(1 - \bar{\sigma}_{\text{eq}})} \right) \\ \langle s | \hat{\eta}_i &= \langle s | \hat{\eta}_i & \langle s | \hat{d}_j^\dagger &= \langle s | \left( \bar{\sigma}_{\text{eq}} + \hat{\eta}_i \sqrt{\bar{\sigma}_{\text{eq}}(1 - \bar{\sigma}_{\text{eq}})} \right) \end{aligned}$$

Obviously, the application of the operator  $\hat{X}$  onto the reference state  $\langle s |$  is equivalent to the application of a corresponding reduced operator  $\hat{X}'$  containing only operators  $\hat{\eta}_k$  (and the trivial operators  $\hat{1} = 1$ ) onto the state  $\langle s |$ , i.e. there is a definitely mapping

$$\hat{X} \Rightarrow \hat{X}' \quad \text{with} \quad \langle s | \hat{X} = \langle s | \hat{X}'$$

Therefore, one obtains (see eq.4):

$$\langle \hat{X} \hat{B}_{\mathbf{N}_n}^{(n)} \rangle = \langle s | \hat{X} \hat{B}_{\mathbf{N}_n}^{(n)} | F \rangle = \langle s | \hat{X}' \hat{B}_{\mathbf{N}_n}^{(n)} | F \rangle = \langle \hat{X}' \hat{B}_{\mathbf{N}_n}^{(n)} \rangle$$

and consequently by using (B1):

$$\langle s | \hat{X} = \langle s | \hat{X}' = \sum_n \sum_{\mathbf{N}_n} \langle \hat{X}' \hat{B}_{\mathbf{N}_n}^{(n)} \rangle \langle s | \hat{B}_{\mathbf{N}_n}^{(n)} = \sum_n \sum_{\mathbf{N}_n} \langle \hat{X} \hat{B}_{\mathbf{N}_n}^{(n)} \rangle \langle s | \hat{B}_{\mathbf{N}_n}^{(n)}$$

i.e. an arbitrary operator  $\hat{X}$  of the Fock-space can be completely presented by using the basis  $\tilde{B} = \{ \hat{B}_{\mathbf{N}_n}^{(n)} \}$  under the consideration that this operator acts into the left direction on the reference state  $\langle s |$ .

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FIG. 1. Arrhenius-plot of the relaxation times  $\tau_R^1(q)$  and  $\tau_R^2(q)$  as a function of the reduced inverse temperature  $\varepsilon/T$ . The full lines correspond to  $q = 0$ , the dashed lines are related to  $q = l^{-1}$  ( $l$  is the unit size of the lattice cells). The dotted line corresponds to an Arrhenius like process. The deviation between the dotted line and the full line indicates a weak non-Arrhenius behavior of  $\tau_R^1(q)$ .

FIG. 2. Intensities  $A_1$  and  $A_2$  as a function of the reduced inverse temperature  $\varepsilon/T$ . The full lines correspond to  $q = 0$ , the dashed lines are related to  $q = l^{-1}$ .

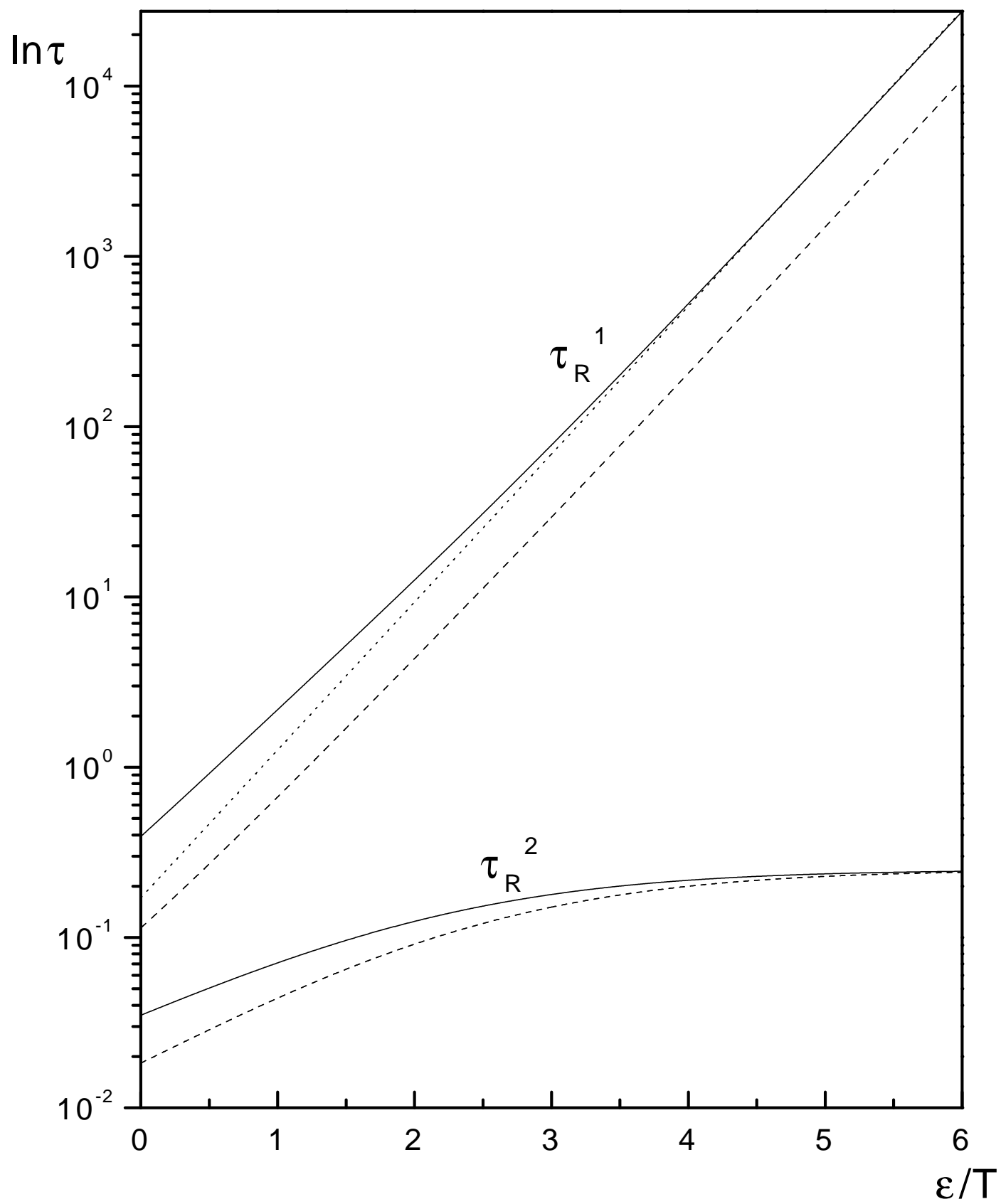
FIG. 3. Correlation function  $\tilde{\Phi}(q, t)$  for various reduced temperatures  $T/\varepsilon = 6.0, 4.0, 2.0, 1.0, 0.5$  and  $0.1$ . The temperature decreases in the direction of the arrow.

FIG. 4. Decay of the reduced correlation function  $\varphi(t)$  for various reduced temperatures  $T/\varepsilon = 0.11, 0.15, 0.22$  and  $0.45$ . The temperature decreases in the direction of the arrow. The functions show with decreasing temperatures an increase of the stretching.

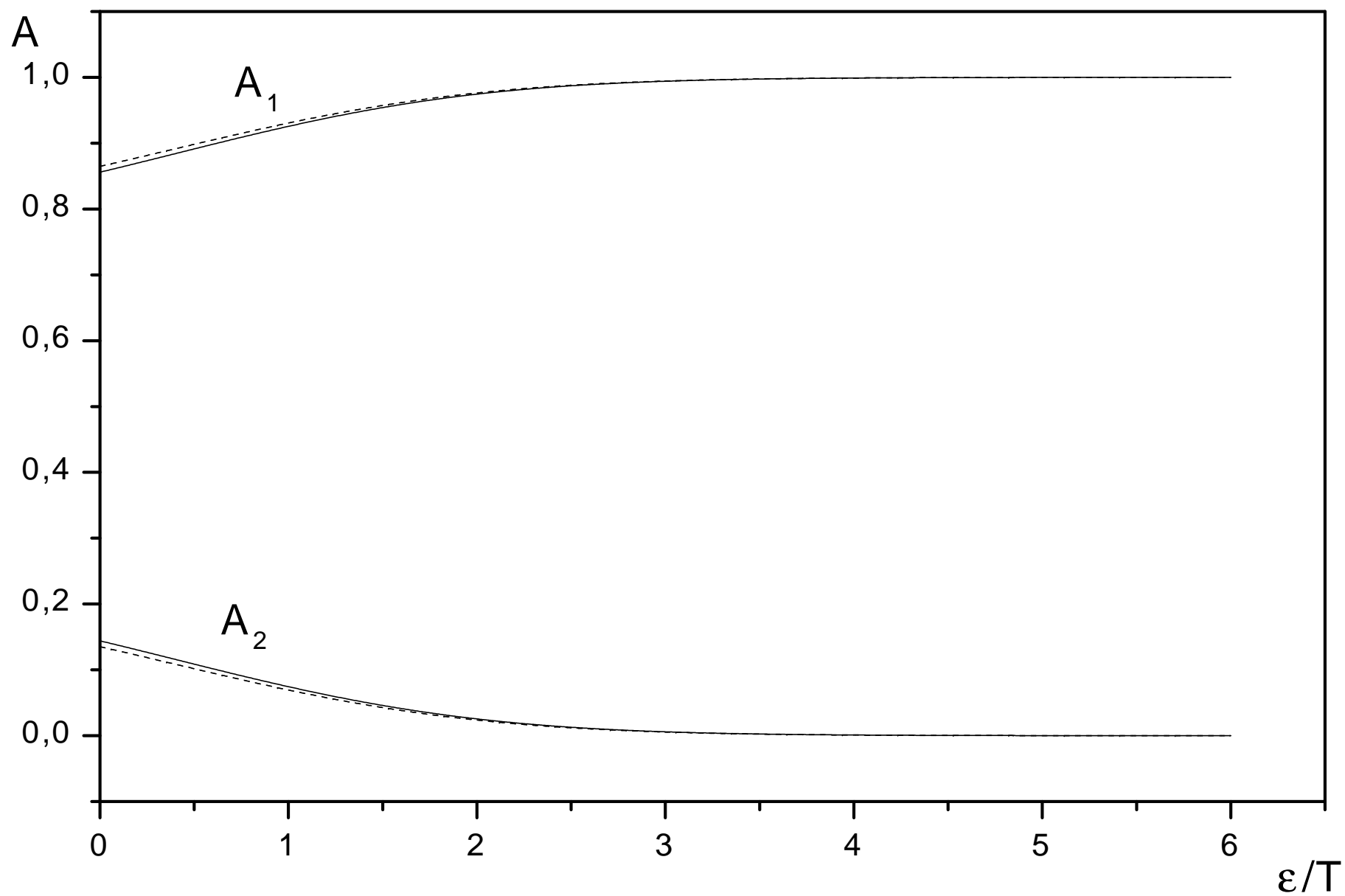
FIG. 5. Spectral density for various  $T/\varepsilon = 0.11, 0.15, 0.22$  and  $0.45$ .

FIG. 6. Suzeptibility  $\chi(\omega)$  for various reduced temperatures  $T/\varepsilon = 0.11, 0.15, 0.22$  and  $0.45$ . The temperature decreases in the direction of the arrow.

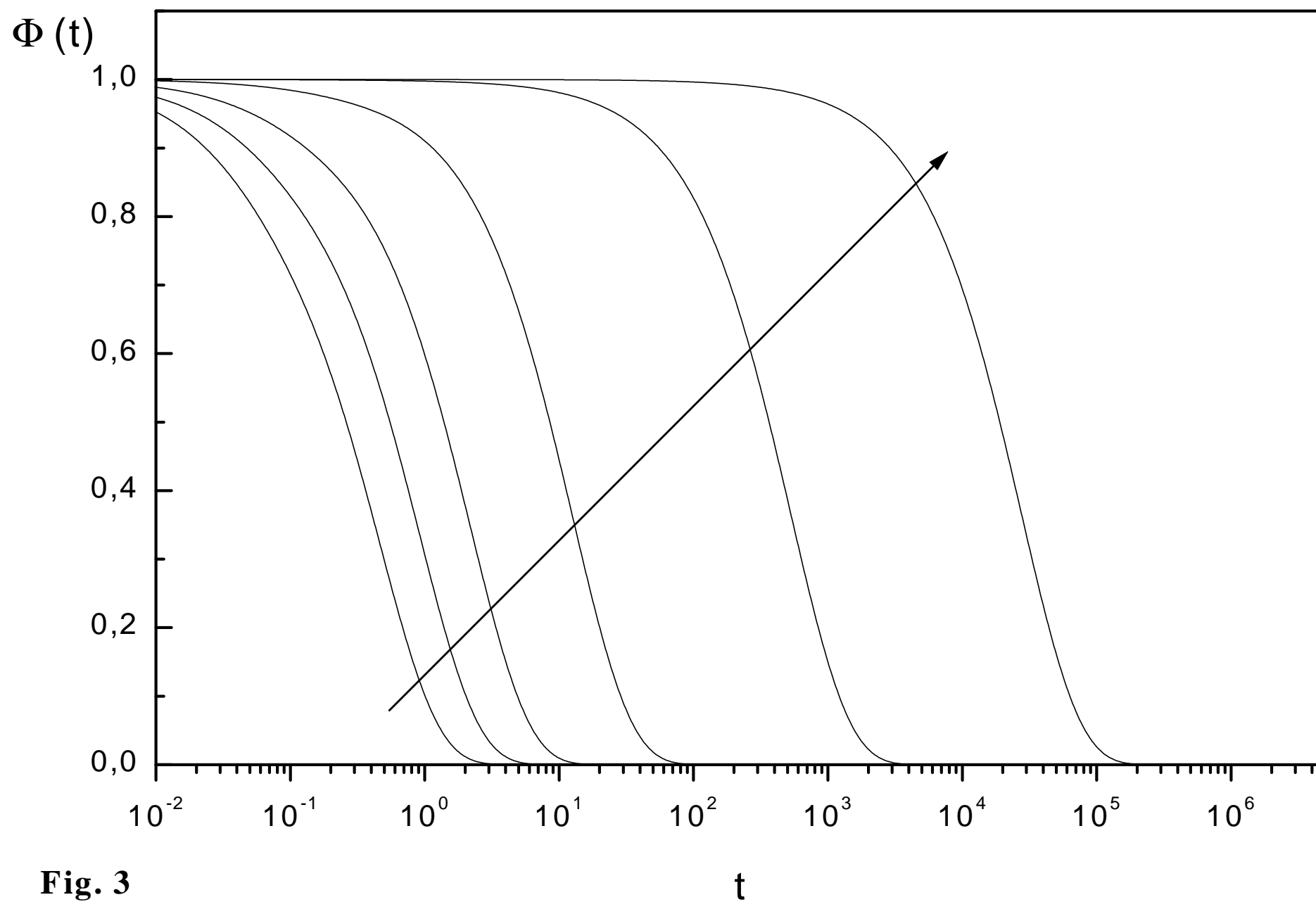
FIG. 7. Relaxation time  $\tau$  as a function of the inverse reduced temperatures  $\varepsilon/T$ .



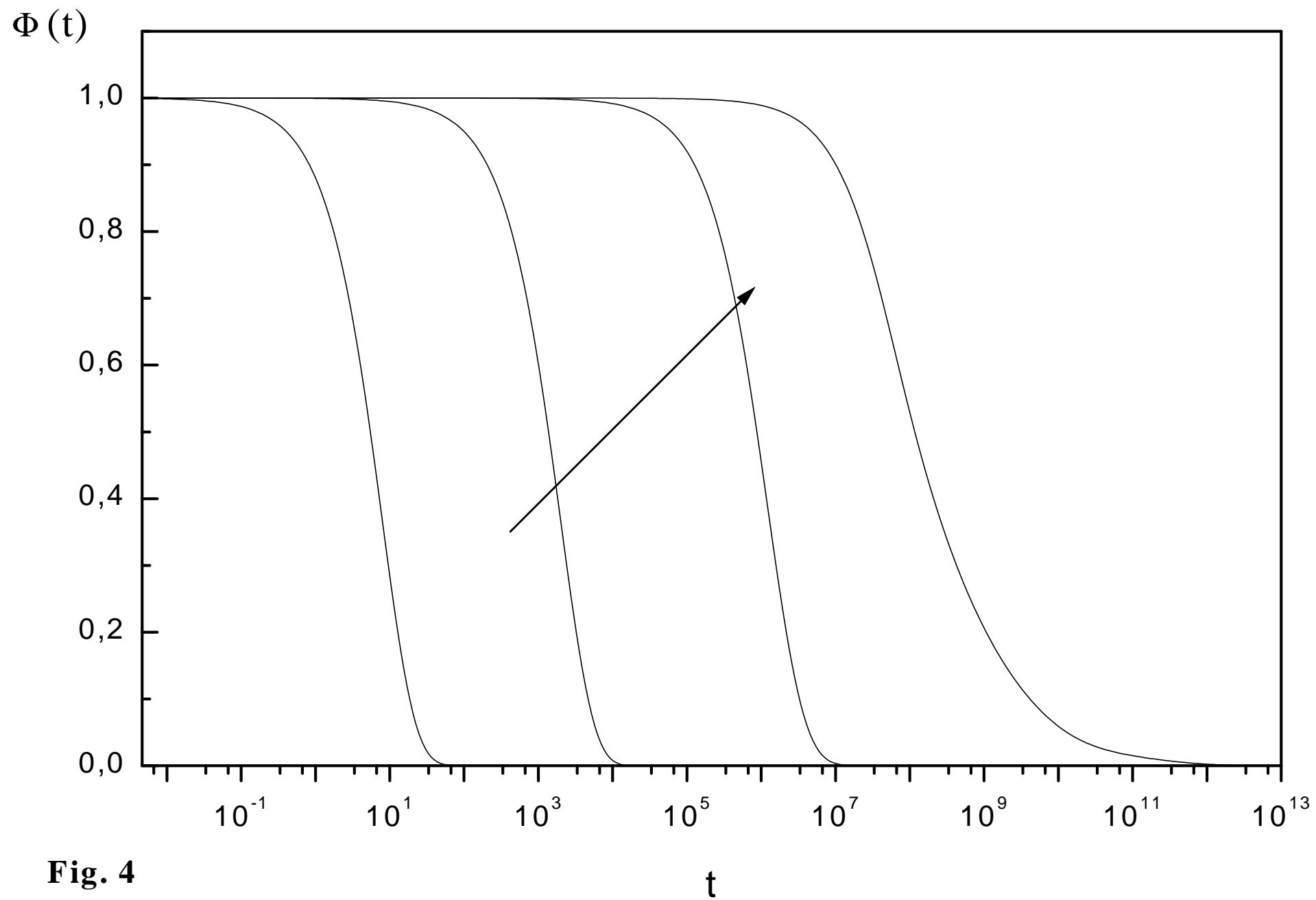
**Fig. 1**



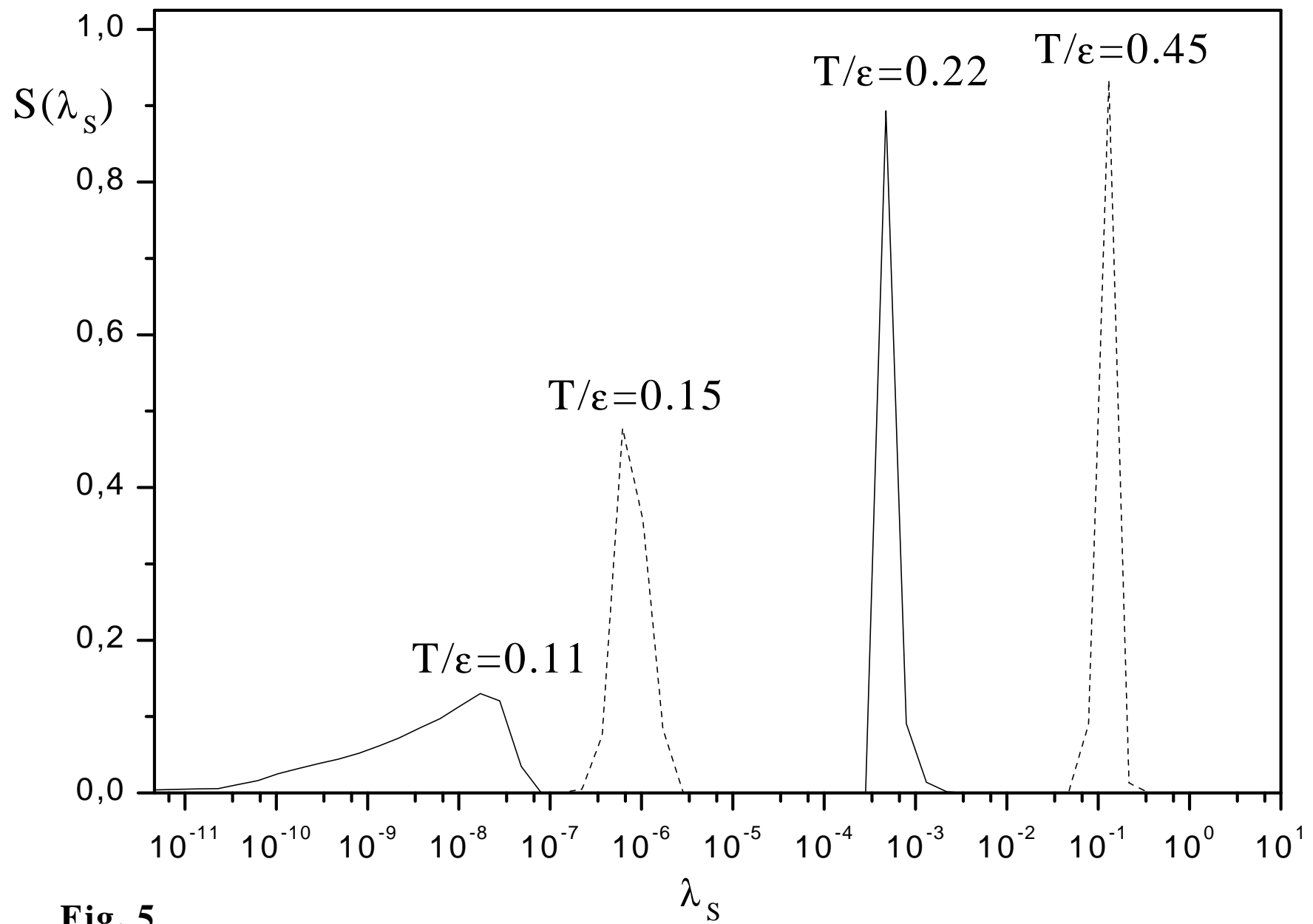
**Fig. 2**



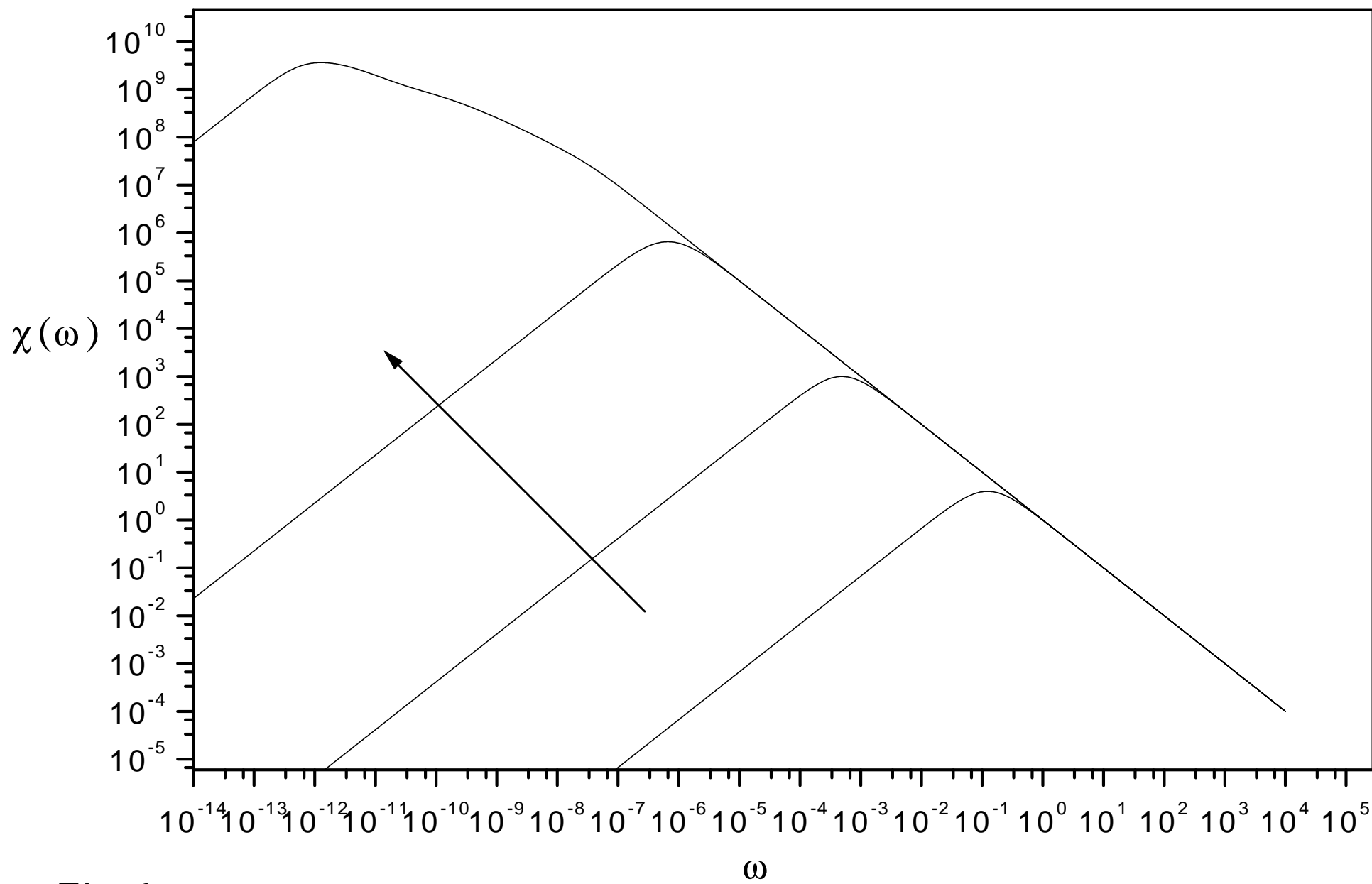
**Fig. 3**



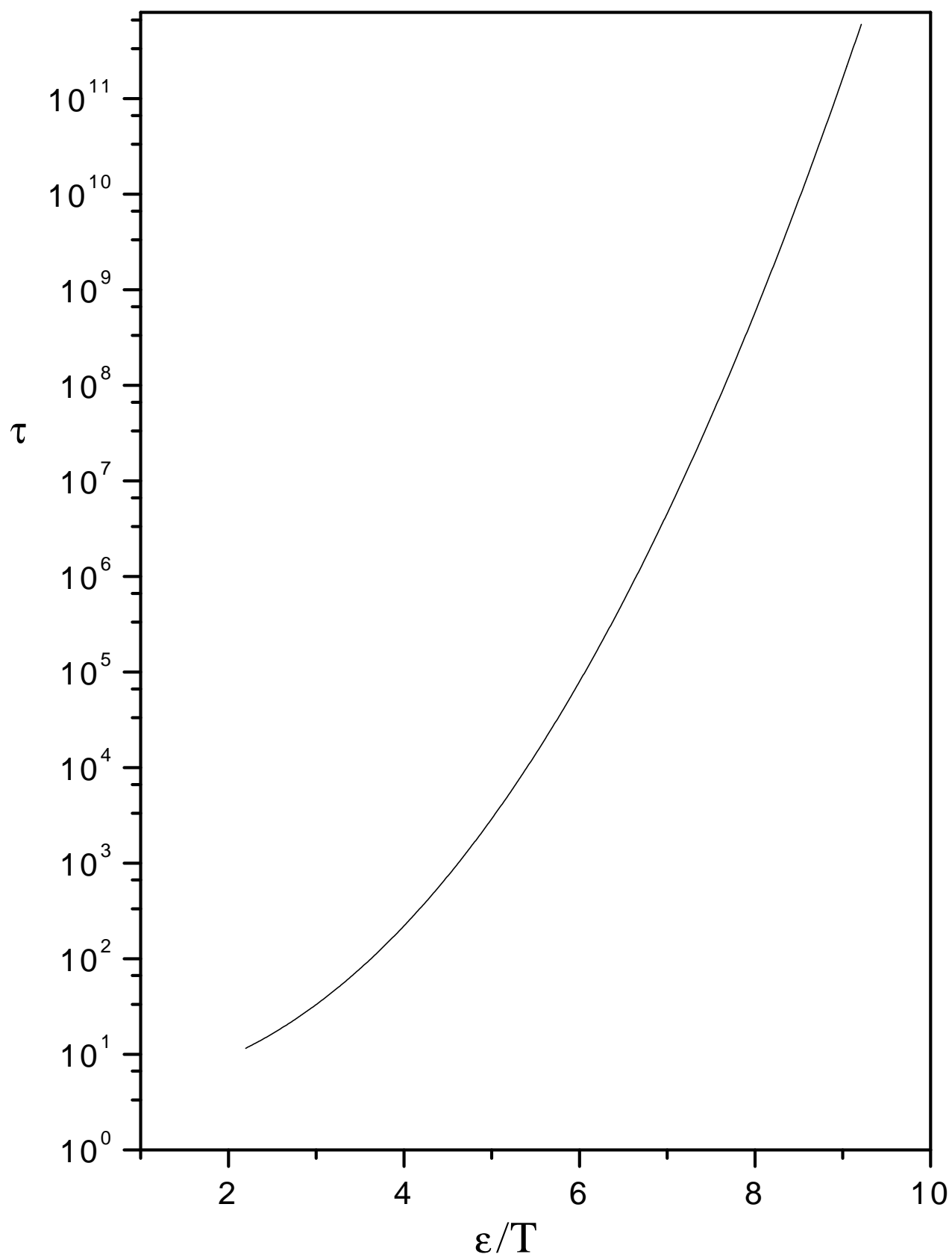
**Fig. 4**



**Fig. 5**



**Fig. 6**



**Fig. 7**